

Evaluation of the diffuse mismatch model for phonon scattering at disordered interfacesQichen Song^{✉*} and Gang Chen^{✉†}*Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

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Diffuse phonon scattering strongly affects the phonon transport through a disordered interface. The often-used diffuse mismatch model assumes that phonons lose memory of their origin after being scattered by the interface. Using mode-resolved atomistic Green's function simulation, we demonstrate that diffuse phonon scattering by a single disordered interface cannot make a phonon lose its memory and thus the applicability of the diffusive mismatch model is limited. An analytical expression for diffuse scattering probability based on the continuum approximation is also derived and shown to work reasonably well at low frequencies.

DOI: [10.1103/PhysRevB.104.085310](https://doi.org/10.1103/PhysRevB.104.085310)**I. INTRODUCTION**

The interface between two dissimilar materials, or even same materials but different crystal orientations, can scatter phonons and gives rise to the thermal boundary resistance for heat flow across the interface [1–3]. One model for the thermal boundary resistance is based on assuming that phonons are specularly scattered at the interface and computing the phonon transmittance and reflectance based on acoustic wave equations, i.e., the acoustic mismatch model (AMM) [2,4]. However, it was found that the AMM only works at very low temperatures at which the phonon wavelengths are long. At elevated temperatures, phonons of short wavelengths carry most of the heat, and they do not experience specular transmission/reflection due to interface imperfections, such as atomic mixing. The diffuse mismatch model (DMM) is proposed as an extreme to describe phonon transport across such rough interfaces [1]. Two major assumptions are made in the DMM. First, the transmittance is isotropic, i.e., transmittance is angle independent. Second, phonons lose memory of their origin after being scattered by the interface such that one cannot distinguish if a phonon has just been through a transmission or reflection process. Although the DMM has improved the agreement with experimental measurements of thermal boundary resistance at high temperatures [5,6], the assumptions behind DMM have never been examined in detail.

The thermal boundary resistance has been studied using equilibrium molecular dynamics [7–9] and nonequilibrium molecular dynamics [10–12]. In particular, phonon-mode-resolved transmittance had been formulated, which builds upon the atomic trajectories at the steady state from molecular dynamics (MD) simulations [13], where the anharmonicity of phonons is intrinsically included. It has been applied to study mode-resolved transmittance through perfect interfaces, yet it has not been used to examine details of diffuse phonon

transmittance across a disordered interface. The phonon wave-packet dynamics technique has been applied to compute the transmittance and reflectance of each phonon mode, and the Kapiza resistance can be calculated using Landauer formalism [14,15]. The MD simulations and, especially, the wave-packet dynamics simulations, however, require large structures in real space, including two bulk regions and the interface region, and simulating phonon transport through a rough interface with a large lateral dimension becomes computationally extensive.

The atomistic Green's function (AGF) has been shown as an effective method to study phonon interfacial transport [16–18]. The method is formulated in reciprocal space and in frequency domain such that one does not have to deal with large-scale simulations of atomic displacements in real space with long simulation time. Recent advances in calculation of interfacial thermal resistance using the AGF have provided more insight into understanding interfacial thermal resistance with detailed information including mode-resolved transmission coefficients [16,19–21]. Specifically, Ong *et al.* studied the phonon specularity and coherence for phonon transport through a disordered grain boundary in two-dimensional graphene and showed that incoherent phonon scatterings at interface are almost perfectly diffusive [22]. Ong also demonstrated that the specular parameters are different for transmittance and reflectance for graphene grain boundaries [23]. Using AGF combined with *ab initio* interatomic force constants, Tian *et al.* found that the intermixing of atoms for Si/Ge interface can enhance interfacial thermal conductance [18]. Sadasivam *et al.* demonstrated using phonon-eigenspectrum-based formulation of AGF that the enhanced interfacial thermal conductance of a Si/Ge interface with atom intermixing comes from diffuse transmission channels where the in-plane momentum is not conserved [20]. The scattering boundary method (SBM), a mathematically equivalent method to AGF, has been proposed by Young and Maris [24] and generalized by Zhao and Freund [25] to study mode-resolved phonon scattering at the interface. Lu and McGaughey [26] have applied SBM to study phonon scattering at the interface between two-dimensional materials. Recently, Latour *et al.* demonstrated the transmission spectra across a

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perfect interface as a function of incident angle of phonons, using mode-resolved AGF [21]. However, to study diffuse phonon scattering, a supercell of a rough interface with a large lateral dimension is required. In addition, the folded lateral wave vector in the supercell must be carefully mapped back to the wave vector defined in the original unit cell. We realize that, despite these studies, none of them had critically examined the validity of DMM for diffuse phonon scattering.

In this paper, we conduct mode-resolved AGF calculation of transmittance and reflectance and revisit the assumptions of DMM. Our study reveals that most phonons do not lose their memory of origin. We also derive an analytical expression for the diffuse transmittance and reflectance based on a continuum model and show that it works reasonably well at low frequencies.

II. METHODOLOGY

A. Revisiting DMM

To derive the DMM, Swartz and Pohl [1] made two major assumptions. The first assumption is that phonons are diffusely scattered by the interface and the transmittance is isotropic,

$$T_{L \rightarrow R}(\omega, \mathbf{q}v) = T_{L \rightarrow R}(\omega), \quad (1)$$

where \mathbf{q} is the wave vector of the phonon with frequency ω and v is the phonon branch index on the left side. The second assumption is that the transmittance from one side must equal the reflectance from the other side, i.e., complete loss of memory:

$$R_{R \rightarrow R}(\omega, \mathbf{q}v) = T_{L \rightarrow R}(\omega). \quad (2)$$

Consequently, the transmittance from right side writes

$$T_{R \rightarrow L}(\omega, \mathbf{q}v) = 1 - T_{L \rightarrow R}(\omega). \quad (3)$$

At a given frequency, by invoking the principle of detailed balance, the transmittance in the elastic scattering limit writes

$$\begin{aligned} T_{L \rightarrow R}(\omega) &= \frac{\sum_{\mathbf{q}v}^+ \frac{v_{Rz, \mathbf{q}v}}{V_{uc, R}} \delta(\omega - \omega_{R, \mathbf{q}v})}{\sum_{\mathbf{q}v}^+ \frac{v_{Lz, \mathbf{q}v}}{V_{uc, L}} \delta(\omega - \omega_{L, \mathbf{q}v}) + \sum_{\mathbf{q}v}^+ \frac{v_{Rz, \mathbf{q}v}}{V_{uc, R}} \delta(\omega - \omega_{R, \mathbf{q}v})} \\ &= \frac{\Theta_{\text{bulk}, R}(\omega)}{\Theta_{\text{bulk}, L}(\omega) + \Theta_{\text{bulk}, R}(\omega)}, \end{aligned} \quad (4)$$

where $v_{\alpha z, \mathbf{q}v}$ with $\alpha = L, R$ is the group velocity normal to the interface for phonons from the left or right side, $\omega_{\alpha, \mathbf{q}v}$ is the phonon frequency and $V_{uc, \alpha}$ is the volume of unit cell of the left and right side. The superscript $+$ means that only forward-moving states with $v_{\alpha z, \mathbf{q}v} > 0$ are included in the summation. The transmittance can also be written in terms of the ratio of transmission functions, as expressed in the second line of the equation. $\Theta_{\text{bulk}, L/R}(\omega)$ is the bulk transmission function for the left/right side, which is a measure of the number of heat conduction channels.

To assess the validity of DMM, we examine if the transmittance and reflectance are indeed isotropic and if the transmittance from one side and reflectance from the other side are the same.

B. Mode-resolved atomistic Green's function formalism

The essential physical quantities to study diffuse phonon scattering by a rough interface are the transmission probability matrix $T_{mn}(\omega)$ and reflection probability matrix $R_{ln}(\omega)$ at a given phonon frequency ω , which describe the transition probability from the initial state n to the final state l or m via interface scattering processes. These matrices are computed from mode-resolved AGF formalism as outlined in Refs. [19,22,27], with details provided in the Supplemental Material [28]. Specifically, we divide the system of interest into three parts, the left lead, the right lead, and the device. We have applied decimation technique in computing the surface Green's function for the leads [29]. The recursive Green's function method is used to compute the Green's function for the device region [30,31]. A propagating state coming out of one lead can be transmitted through (or be reflected by) the device region and travel to the other lead (or the same lead). We then compute the ratio of the heat flux along the z direction of the outgoing state m (or state l) to the heat flux along the z direction of the initial state n , which is the element of transmission probability matrix $T_{mn}(\omega)$ [or reflection probability matrix $R_{ln}(\omega)$].

For a rough interface created by atomic mixing at the interface, the transverse translational symmetry is broken by the interfacial disorders. It is impractical to compute scatterings of an infinitely large rough interface. Instead, we construct a supercell of two materials and a rough interface between them with periodic boundary conditions along the transverse directions (x direction and y direction). Because of the transverse periodicity of the supercell, the phonon state of the lead region defined at a given transverse wave vector $\mathbf{q}_{\text{sc}, \parallel}$ can only be scattered into phonon states of the lead region (either the left or the right lead) with the same wave vector $\mathbf{q}_{\text{sc}, \parallel}$.

The lead part of the supercell contains $N_x \times N_y$ repeated unit cells, as depicted in Fig. 1(a). The period lengths of the lead along the direction normal to the interface are $a_{z, L}$ for the left lead and $a_{z, R}$ for the right lead. The phonon wave vectors parallel to the interface in the supercell and in the unit-cell representations are related via $\mathbf{q}_{\text{uc}, \parallel} = \mathbf{q}_{\text{sc}, \parallel} + a\mathbf{G}_{\text{sc}, x} + b\mathbf{G}_{\text{sc}, y}$, where a and b are integers. $\mathbf{G}_{\text{sc}, x}$ and $\mathbf{G}_{\text{sc}, y}$ are transverse reciprocal lattice vectors of the supercell. The phonon states at the corresponding equivalent wave vectors (with same q_z and same branch index) in the two representations are equivalent [32]. The phonon state in the unit-cell representation is preferred as it is much easier to interpret than the supercell representation (we will hide subscript uc in the following for visual clarity). However, for a given supercell state $\mathbf{q}_{\text{sc}, \parallel}$, there are multiple possible choices of a and b . To find out the correct pair of a and b for wave vector \mathbf{q}_{\parallel} is known as an unfolding problem. We have adopted the unfolding scheme by Popescu and Zunger [33] and the details can be found in the Supplemental Material [28].

For an interface scattering event, the transverse wave vectors for initial state n and final state m in the unit-cell representation are constrained by

$$\mathbf{q}_n = \mathbf{q}_m + a\mathbf{G}_{\text{sc}, x} + b\mathbf{G}_{\text{sc}, y}, \quad (5)$$

where a and b are unknown integers, as wave vectors of initial and final states in the supercell representation can be unfolded

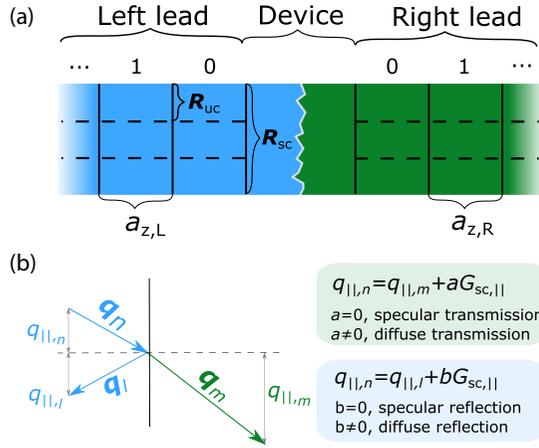


FIG. 1. (a) In AGF, the system is partitioned into three parts: two semi-infinite leads and a rough interface as the device region. The lead region in the supercell contains N_{uc} unitcells and transverse lattice vector $\mathbf{R}_{sc} = N_{uc}\mathbf{R}_{uc}$. The numbers 0, 1, ... denote the index of repeated cells for left and right leads. The period lengths along the direction normal to the interface are $a_{z,L}$ and $a_{z,R}$ for the left and right leads, respectively. (b) The in-plane wave vector for incident, transmitted, and reflected phonons, $\mathbf{q}_{||,n}$, $\mathbf{q}_{||,m}$, and $\mathbf{q}_{||,l}$. $a = 0$ corresponds to specular transmission, while $a \neq 0$ corresponds to diffuse transmission. Similarly, $b = 0$ corresponds to specular reflection, while $b \neq 0$ corresponds to diffuse reflection. Note the schematic is drawn for two-dimensional system for visual clarity and for three-dimensional system the partitioning and wave-vector conservation laws can be analogously defined.

differently. This expression indicates that the interface scattering can either be a momentum conserved (specular), when $a = b = 0$, or momentum nonconserved (diffuse) process for other a and b values. Depending on the transverse wave vector of state n and state m , the transmission probability matrix can be categorized into specular and diffuse transmission parts $T_{mn}(\omega) = T_{s,mn}(\omega) + T_{d,mn}(\omega)$, where

$$\begin{cases} T_{s,mn}(\omega) = T_{mn}(\omega), & \text{when } \mathbf{q}_{||,n} = \mathbf{q}_{||,m} \\ T_{d,mn}(\omega) = T_{mn}(\omega), & \text{when } \mathbf{q}_{||,n} \neq \mathbf{q}_{||,m}. \end{cases} \quad (6)$$

The reflection probability matrix can be analogously expressed by $R_{ln}(\omega) = R_{s,ln}(\omega) + R_{d,ln}(\omega)$, where

$$\begin{cases} R_{s,ln}(\omega) = R_{ln}(\omega), & \text{when } \mathbf{q}_{||,n} = \mathbf{q}_{||,l} \\ R_{d,ln}(\omega) = R_{ln}(\omega), & \text{when } \mathbf{q}_{||,n} \neq \mathbf{q}_{||,l}. \end{cases} \quad (7)$$

The diffuse transmittance for a given incident phonon n from the left side is defined by summing over the scattering probabilities of all possible outgoing states,

$$T_{d,L \rightarrow R}(\omega, \Omega_L) = \sum_m T_{d,mn}^{L \rightarrow R}(\omega), \quad (8)$$

where $\Omega_L = (\theta, \phi)$ indicates transmittance is a directional quantity. The polar and azimuthal angles are defined in a coordinate system where the interface normal lies along the z axis, $\theta = \arccos \frac{v_z}{|\mathbf{v}|}$, $\phi = \arctan \frac{v_y}{v_x}$. $\mathbf{v} = (v_x, v_y, v_z)$ is the group velocity for incident phonon n from the left side and we use $L \rightarrow R$ to denote the trajectory of the phonon. The reason for using the angle of the group velocity rather than the phase velocity (or wave vector) is that group velocity is a uniquely

defined quantity irrelevant to the choice of in-plane Brillouin zone while not for the phase velocity.

Likewise, the diffuse reflectance for a given initial state n from the left side and from the right side read

$$R_{d,\alpha \rightarrow \alpha}(\omega, \Omega_\alpha) = \sum_m R_{d,mn}^{\alpha \rightarrow \alpha}(\omega), \quad (9)$$

where $\alpha = L, R$. The specular transmittance and reflectance can be similarly defined.

Furthermore, to study the impact of diffuse phonon scattering on interfacial transport, we compute the transmission function $\Theta(\omega)$, which accounts for the total phonon conduction transmission at a given frequency ω , defined by

$$\Theta(\omega) = \Theta_s(\omega) + \Theta_d(\omega). \quad (10)$$

$\Theta_s(\omega)$ and $\Theta_d(\omega)$ are specular and diffuse transmission function obtained by summing over all possible incoming and outgoing states at a given frequency [34]:

$$\begin{aligned} \Theta_s(\omega) &= \sum_{mn} T_{s,mn}^{L \rightarrow R}(\omega), \\ \Theta_d(\omega) &= \sum_{mn} T_{d,mn}^{L \rightarrow R}(\omega). \end{aligned} \quad (11)$$

The specular and diffuse reflection function is defined by

$$\begin{aligned} \Xi_{s,\alpha}(\omega) &= \sum_{mn} R_{s,mn}^{\alpha \rightarrow \alpha}(\omega), \\ \Xi_{d,\alpha}(\omega) &= \sum_{mn} R_{d,mn}^{\alpha \rightarrow \alpha}(\omega), \end{aligned} \quad (12)$$

with $\alpha = L, R$. Note that the transmission function for two sides are the same due to the time-reversal symmetry of the transmission probability matrix but not for the reflection function.

C. Continuum modeling

In addition to the AGF simulation, we also derived analytical formulas for diffuse transmittance and reflectance from continuum modeling with details provided in the Appendix. The model assumes scalar acoustic waves and random mass disorders distributed at the interface, and hence neglects mode conversion at the interface. The model is derived based on perturbation theory and it only takes the densities ρ_L, ρ_R , bulk moduli μ_L, μ_R , and number of pairs of swapped atoms at the interface per unit area n as parameters. The model captures the specific contributions to the total transmittance/reflectance of specular and diffuse scattering processes and allows one to calculate diffuse/specular transmittance and reflectance components analytically.

We assume a linear dispersion $\omega = c|\mathbf{q}|$, where the sound velocities for the left and right sides are $c_L = \sqrt{\mu_L/\rho_L}$ and $c_R = \sqrt{\mu_R/\rho_R}$, respectively. Due to the simple dispersion relation, the transverse $\mathbf{q}_{||}$ can uniquely define a forward-moving phonon state. Thus, we use $\mathbf{q}_{||}$ and $\mathbf{q}'_{||}$ to denote the initial and final states, instead of using m and n . The momentum for a phonon state on the left side is $(\mathbf{q}_{||}, q_L) = \frac{\omega}{c_L}(\sin\theta_L \cos\phi, \sin\theta_L \sin\phi, \cos\theta_L)$, where q_L is the perpendicular momentum. For a specular transmission process from the

left side to the right side, the transverse momentum is conserved. Thus, the corresponding transmitted phonon state on the right side is $(\mathbf{q}_{\parallel}, q_R) = \frac{\omega}{c_R}(\sin\theta_R \cos\phi, \sin\theta_R \sin\phi, \cos\theta_R)$. It follows that the perpendicular velocities for the initial and final state are $v_L = c_L \cos\theta_L = c_L \sqrt{1 - c_L^2 |\mathbf{q}_{\parallel}|^2 / \omega^2}$, $v_R = c_R \cos\theta_R = c_R \sqrt{1 - c_R^2 |\mathbf{q}_{\parallel}|^2 / \omega^2}$, respectively.

From our continuum model, the diffuse transmittance for a given incident state from the left side writes

$$T_{d,L \rightarrow R}(\omega, \Omega_L) = \int \frac{d^2 \mathbf{q}'_{\parallel}}{(2\pi)^2} T_d^{L \rightarrow R}(\omega, \mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}), \quad (13)$$

where

$$T_d^{L \rightarrow R}(\omega, \mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}) = 4\omega^{-2} V_2 \frac{\rho_R v'_R}{|\rho_L v'_L + \rho_R v'_R|^2} \frac{\rho_L v_L}{|\rho_L v_L + \rho_R v_R|^2}. \quad (14)$$

Here, V_2 is a quantity related to density of mixed atoms at interface (see Appendix). Essentially, we have integrated over all possible final states \mathbf{q}'_{\parallel} on the right-hand side. Note that the final transverse momentum is bounded by $|\mathbf{q}'_{\parallel}| \leq \frac{\omega}{c_R}$.

The diffuse reflectance for a phonon state from the left-hand side is

$$R_{d,L \rightarrow L}(\omega, \Omega_L) = \int \frac{d^2 \mathbf{q}'_{\parallel}}{(2\pi)^2} R_d^{L \rightarrow L}(\omega, \mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}), \quad (15)$$

where

$$R_d^{L \rightarrow L}(\omega, \mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}) = 4\omega^{-2} V_2 \frac{\rho_L v'_L}{|\rho_L v'_L + \rho_R v'_R|^2} \frac{\rho_L v_L}{|\rho_L v_L + \rho_R v_R|^2}, \quad (16)$$

and the transverse momentum of the final state is bounded by $|\mathbf{q}'_{\parallel}| \leq \frac{\omega}{c_L}$.

If we denote $F(\omega) = \int \frac{d^2 \mathbf{q}'_{\parallel}}{(2\pi)^2} 4\omega^{-2} V_2 \frac{\rho_L v'_L}{|\rho_L v'_L + \rho_R v'_R|^2}$, the diffuse transmittance from one side and the diffuse reflectance from the other side can be, respectively, expressed by

$$T_{d,R \rightarrow L}(\omega, \Omega'_R) = \frac{\rho_R v'_R}{|\rho_L v'_L + \rho_R v'_R|^2} F(\omega), \quad (17)$$

$$R_{d,L \rightarrow L}(\omega, \Omega_L) = \frac{\rho_L v_L}{|\rho_L v_L + \rho_R v_R|^2} F(\omega). \quad (18)$$

It is evident that they are both anisotropic since they depend on perpendicular incident velocity. Their ratio writes

$$\frac{T_{d,R \rightarrow L}(\omega, \Omega'_R)}{R_{d,L \rightarrow L}(\omega, \Omega_L)} = \frac{\rho_R v'_R}{\rho_L v_L} \frac{|\rho_L v_L + \rho_R v_R|^2}{|\rho_L v'_L + \rho_R v'_R|^2}, \quad (19)$$

which is not a constant. Thereby, our continuum model suggests that the diffuse transmittance from one side and the diffuse reflectance from the other side are generally not equal.

III. RESULTS AND DISCUSSIONS

We study phonon transport through a disordered [001] Si/Ge interface by creating a 3×3 supercell (along x and y directions). We use the average of Si's and Ge's lattice constants, $a = 5.527 \text{ \AA}$, as the lattice constant in generating the supercell structures and the Stillinger-Weber interatomic potential to compute the dynamical matrix and Green's function [35]. The disordered interface is constructed by randomly

swapping Si and Ge atoms with the same distances to the interface and the further away from the interface the fewer atoms are swapped. At even further distances from the interface, no Si and Ge atoms are swapped. For instance, when we have two layers of Si and two layers of Ge atoms mixed, two pairs of Si and Ge atoms will be swapped in the Si and the Ge layer closest to the interface and one pair of Si and Ge atoms will be swapped in the Si and the Ge layer second closest to interface. In this case, we have a 1|2|2|1 configuration, with each number denoting the number of swapped atoms within the same layer. We label such interface structure by 4 ml, in short for four mixing layers in total. Apparently, a larger ml number means a larger degree of disorder. We generate 21 configurations for each given total mixing layers and compute the ensemble average of the transmission and reflection probability matrices $T_{mn}^{L \rightarrow R}(\omega)$, $T_{mn}^{R \rightarrow L}(\omega)$, $R_{mn}^{L \rightarrow L}(\omega)$ and $R_{mn}^{R \rightarrow R}(\omega)$.

We first examine the angular dependence of ensemble-averaged diffuse transmittance and reflectance at a given frequency to ascertain whether or not they are isotropic. In Fig. 2, we find strong angle dependence of diffuse transmittance and diffuse reflectance from the Si side as well as the Ge side, contradicting the assumption of isotropic reflectance and transmittance underlying the DMM. The diffuse reflectance from Ge to Ge is found to be overall higher than the diffuse transmittance from Si to Ge. The diffuse reflectance from Si to Si is in a similar range compared with the diffuse transmittance from Ge to Si, although their explicit angle dependences are drastically different.

If the DMM is valid, the diffuse transmittance from one side and the diffuse reflectance from the other side should be isotropic. We compute the difference between the maximum and minimum scattering probability at different frequencies, as a measure of anisotropy, shown in Figs. 2(e) and 2(f). We observe that the diffuse scattering probability generally varies in a wide range. For example, the diffuse reflectance from the Si side at high frequencies ranges almost from zero to one. Hence, we conclude the diffuse transmittance and reflectance from both sides are highly anisotropic.

Additionally, we find out that the patterns of diffuse transmittance and diffuse reflectance do not have two perfect diagonal reflection axes as the perfect Si/Ge [001] interface structure does. This is the consequence of ensemble average of disordered structures, where each structure might have broken the reflection symmetry, and it is not guaranteed to recover the original symmetry after ensemble average. Still, we can clearly observe a cloverlike pattern for the diffuse transmittance and reflectance from the Si side, which originates from the pmm symmetry of the structure for a perfect [001] Si/Ge interface.

In particular, to quantitatively study how the diffuse transmittance and diffuse reflectance depend on the polar angle, we integrate the angle-dependent scattering probability over the azimuthal angle. As shown in Fig. 3(a), at $\omega = 3.3 \text{ THz}$, the diffuse transmittance from Si is lower than the reflectance from Ge, while in Fig. 3(b), the diffuse transmittance from Ge is higher than the diffuse reflectance from Si. It is interesting to note that the continuum model agrees well with the results from AGF when incident states are from Si, suggesting the model captures the physics of diffuse phonon scattering. On the contrary, the continuum model predicts a different diffuse

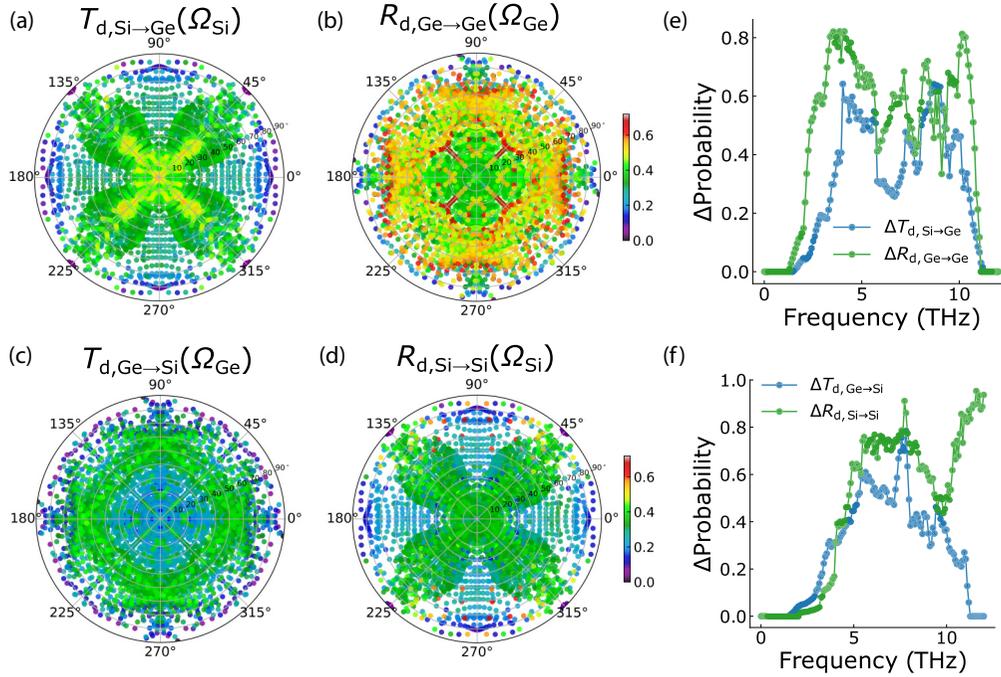


FIG. 2. The angle-resolved (a) diffuse transmittance $T_{d,Si \rightarrow Ge}(\Omega_{Si})$ from the Si side and (b) diffuse reflectance $R_{d,Ge \rightarrow Ge}(\Omega_{Ge})$ from the Ge side at $\omega = 5$ THz. The angle-resolved (c) diffuse transmittance $T_{d,Ge \rightarrow Si}(\Omega_{Ge})$ from the Ge side and (d) diffuse reflectance $R_{d,Si \rightarrow Si}(\Omega_{Si})$ from the Si side at $\omega = 5$ THz. $\Omega_\alpha = (\theta_\alpha, \phi_\alpha)$, $\alpha = \text{Si, Ge}$ is the direction of incident group velocity. The radial coordinate corresponds to the polar angle θ_α (angle of incidence) and the polar axis corresponds to the azimuthal angle ϕ_α . (e), (f) The difference between maximum and minimum diffuse scattering probability as a measure of the anisotropy of diffuse scattering probability. The diffuse scattering probability is obtained by taking the ensemble average of calculations for 21 structures of 8 ml disordered configurations with a 20×20 $\mathbf{q}_{sc,\parallel}$ -point mesh.

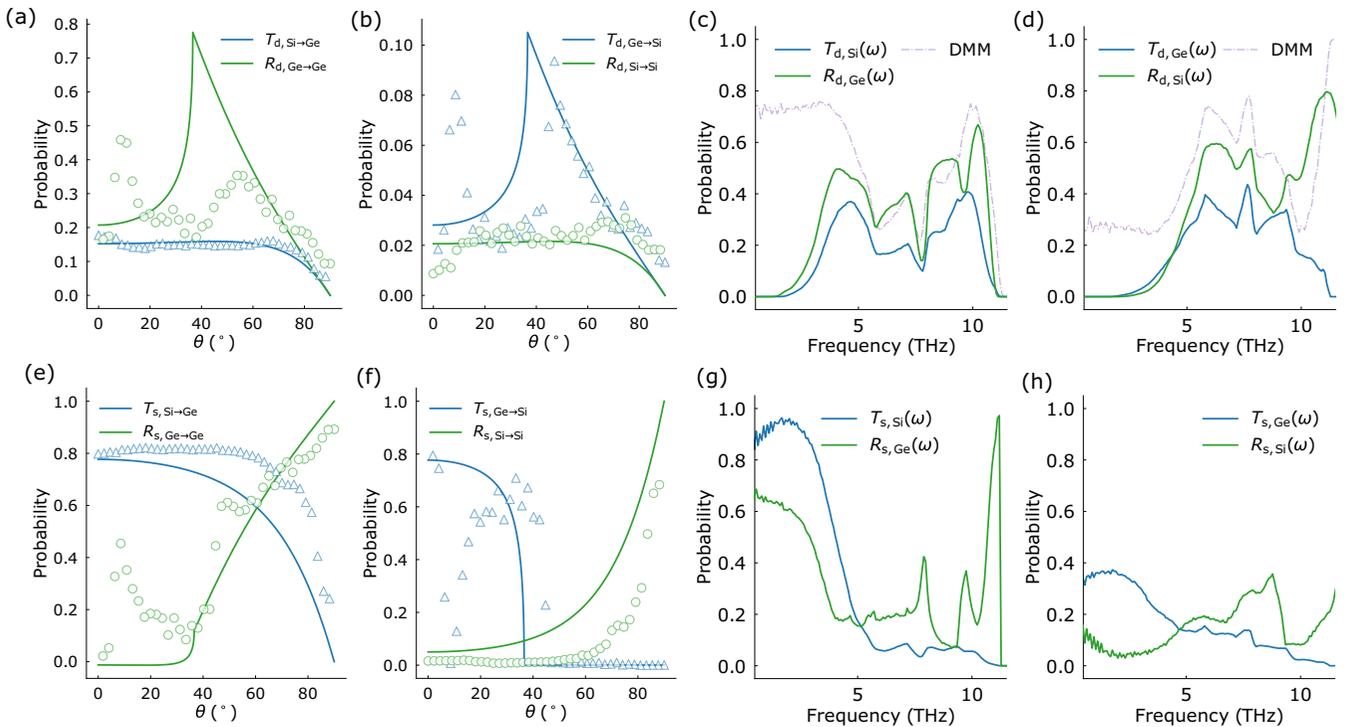


FIG. 3. (a), (b) The diffuse transmittance $T_{d,\alpha}$ from one side and the diffuse reflectance $R_{d,\beta}$ from the other side as a function of the polar angle of the incident phonon at $\omega = 3.3$ THz. The markers are obtained by integrating $T_d(\Omega)$ and $R_d(\Omega)$ from AGF calculation for 8 ml structures over the azimuthal angle ϕ divided by 2π . The solid lines are predictions from continuum modeling with the number of pairs of swapped atoms per unit area $n = 2.78/a^2$ and $a = 5.527$ Å. (c), (d) The average diffuse transmittance $T_{d,\alpha}(\omega)$ from one side and the average diffuse reflectance $R_{d,\beta}(\omega)$ from the other side as a function of frequency from AGF calculation in solid lines, compared with DMM in dash-dot lines. (e)–(h) are the specular transmittance and specular reflectance corresponding to (a)–(d).

scattering probability profile compared with the calculation from AGF when incident phonons are from Ge. From the analytical expression in the continuum model for diffuse scattering probability [Eqs. (13) and (15)], we learn that the peak in diffuse transmittance and reflectance profile corresponds to the critical angle for total internal reflection (37°). In comparison, the scattering probability profile from AGF has two peaks (9° and around 50°). In Figs. 3(e) and 3(f), we find that the specular transmittance and reflectance have a much stronger dependence on the polar angle compared with their diffuse counterparts. Our continuum model for specular scattering probabilities again shows good agreement with the AGF calculation except incapable of capturing the multiple peaks arising from total internal reflection. Note that the continuum modeling is based on a scalar field, where the polarization vectors are not included. The difference between continuum modeling and AGF calculation suggests that without considering mode conversion among different polarizations, the scalar continuum model cannot accurately describe the actual number of available diffuse transmission and reflection pathways.

To study the diffuse transmittance at different frequencies, we define an average diffuse transmittance for phonon modes with the same frequency by

$$T_{d,\alpha}(\omega) = \frac{\sum_n^+ T_{d,n}^{\alpha \rightarrow \beta}(\omega) v_{z,n}}{\sum_n^+ 1 \cdot v_{z,n}} = \frac{\Theta_d(\omega)}{\Theta_{\text{bulk},\alpha}(\omega)}, \quad (20)$$

where $v_{z,n}$ is the group velocity normal to the interface. $\Theta_{\text{bulk},\alpha}(\omega)$ is the transmission function for bulk material α (we use two leads and device all consisting of α atoms in AGF calculation). The average total transmittance including specular transmittance and diffuse transmittance is defined by $T_\alpha(\omega) = \frac{\Theta(\omega)}{\Theta_{\text{bulk},\alpha}(\omega)}$. The average diffuse and total reflectance are $R_{d,\alpha}(\omega) = \frac{\Xi_{d,\alpha}(\omega)}{\Theta_{\text{bulk},\alpha}(\omega)}$ and $R_\alpha(\omega) = \frac{\Xi_\alpha(\omega)}{\Theta_{\text{bulk},\alpha}(\omega)}$, respectively. Note that the sum of transmittance and reflectance is one, yet the sum of diffuse transmittance and diffuse reflectance is less than one, as not all phonons are diffusely scattered. On the other hand, the sum of transmittance and reflectance given by DMM is always unity. Therefore, when we compare the diffuse transmittance/reflectance with transmittance/reflectance by DMM, it is entirely possible their values do not match. However, what we are more interested in answering is whether or not the phonon loses its memory, i.e., whether the diffuse transmittance from one side equals the diffuse reflectance from the other side [36].

In Fig. 3(c), we find that the diffuse transmittance from Si is lower than the diffuse reflectance from Ge for all frequencies. They both deviate from DMM given by Eq. (4) at low frequencies. At high frequencies, the DMM's prediction is close to the reflectance from Ge. In Fig. 3(d), the diffuse transmittance from Ge is lower than reflectance from Si except for low frequencies. And they are both different from DMM. The crossing point at 4.4 THz for transmittance and reflectance suggest that at this frequency, the average transmittance from the Ge side is the same with average diffuse reflectance from the Si side, although they individually have strong angle dependence. The gap between diffuse transmittance from one side and diffuse reflectance from the other side suggests that diffuse phonon scattering depends on the initial state such that phonons actually do not lose their

memory of origin. In Figs. 3(g) and 3(h), we see that the specular scattering probability is generally much higher than the diffuse scattering probability at low frequencies, suggesting that at low frequencies, the interface scattering is almost all specular. This trend is also consistent with previous findings [20,37].

An important factor that is relevant to the diffuse phonon scattering is the amount of dissimilarity between two materials. According to Swartz and Pohl [1], DMM predicts that diffuse scattering increases thermal boundary resistance of the interface between similar solids, suggesting that diffuse scattering plays a significant role when the mass ratio between the two materials is close to one. In Fig. 4, we present the frequency-resolved transmittance and reflectance with different mass ratios $m_{\text{Ge}}/m_{\text{Si}}$ (we fix the mass m_{Si} and vary the mass m_{Ge}). We find that when the two sides are similar, the diffuse transmittance from one side is similar to the diffuse reflectance from the other side. This partially aligns with the assumptions of DMM, although the diffuse transmittance and diffuse reflectance still have strong anisotropy in directions (see Fig. 6 in the Supplemental Material [28]). When the mass ratio is large, the transmittance from one side is no longer similar to the reflectance from the other side, indicating that the scattering probability strongly depends on where the initial states are from. As for the total transmittance and reflectance, due to the inclusion of the specular scattering probability, the difference between transmittance from one side and reflectance from the other side is enlarged. It is interesting to note that when two sides are similar, although DMM cannot correctly describe either scattering probability, it is close to the average of the total transmittance from one side and the total reflectance from the other side. When the two sides are dissimilar, the DMM's prediction becomes similar to the total transmittance in Ge side at low frequencies and the total transmittance in Si side at high frequencies. This suggests that for certain cases, DMM is able to roughly describe the total transmittance from one side in a certain frequency range but not for all frequencies.

To study how much memory the phonon loses regarding its origin in a quantitative manner, we define a similarity measure by

$$S_\beta(\omega) = \exp\left(-\frac{|T_{d,\alpha}(\omega) - R_{d,\beta}(\omega)|}{R_{d,\beta}(\omega)}\right), \quad (21)$$

where $T_\alpha(\omega)$ is the diffuse transmittance from one side and $R_\beta(\omega)$ is the diffuse reflectance from the other side, and the final states for these scattering processes are on the β side. $S_\beta \rightarrow 1$ means high similarity between the diffuse transmittance and reflectance when the final state is on the β side, i.e., phonon completely loses its memory of origin. $S_\beta \rightarrow 0$ means low similarity between the diffuse transmittance from one side and diffuse reflectance from the other side, and phonon does not lose its memory. A large mass ratio generally lead to a smaller similarity measure. We also find that the similarity measure depends which side the final state resides at. For example, in Fig. 5(b), we find that at $\omega = 0.39\omega_{\text{max}}$, when a phonon is scattered into the Si side, it loses its memory. However, this is not true when a phonon is scattered into the

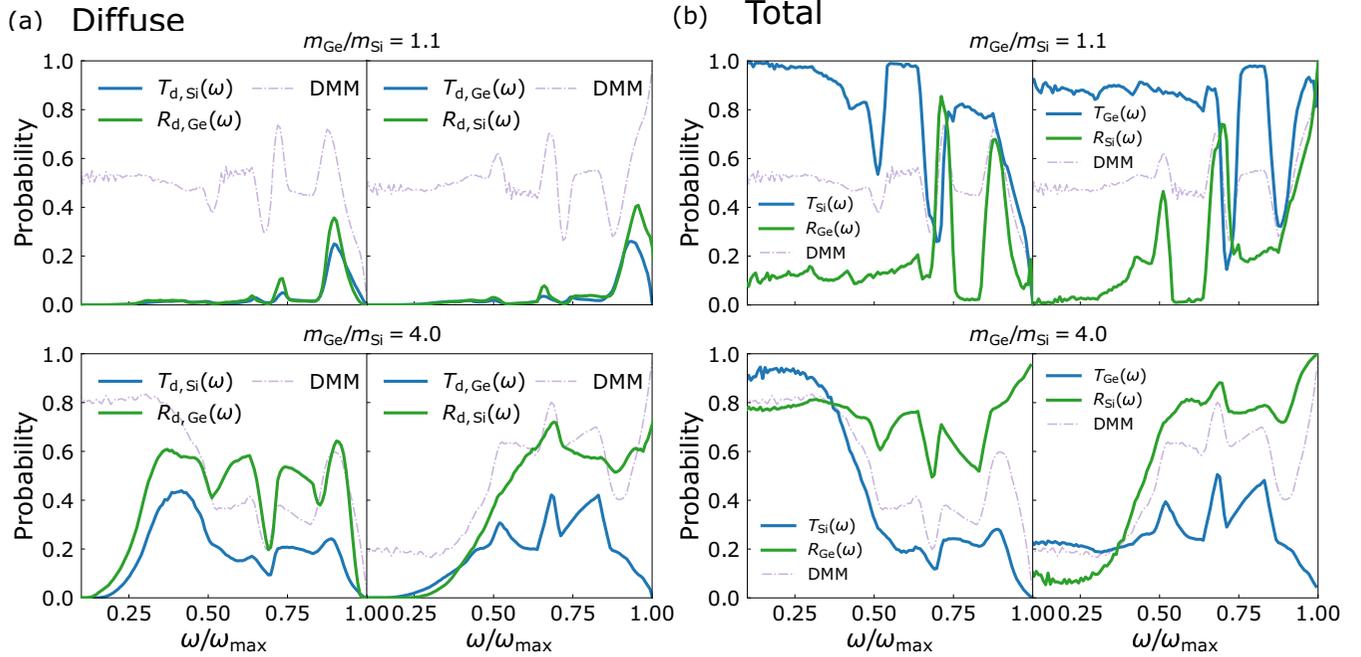


FIG. 4. The frequency-resolved (a) diffuse and (b) total transmittance/reflectance from Si and Ge side with different $m_{\text{Ge}}/m_{\text{Si}}$ from AGF calculation for 8-ml structures and DMM. ω_{max} is the maximum allowed frequency for nonzero transmission function $\Theta(\omega)$. $\omega_{\text{max}} = 17.2$ THz when $m_{\text{Ge}}/m_{\text{Si}} = 1.1$, and $\omega_{\text{max}} = 9.1$ THz when $m_{\text{Ge}}/m_{\text{Si}} = 4.0$.

Ge side. In comparison, for DMM, the similarity is always one when the final states are on either side of the interface.

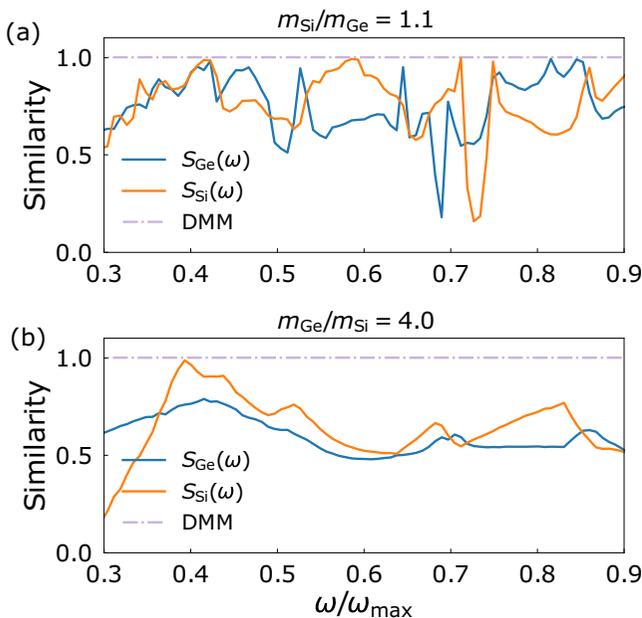


FIG. 5. The similarity between the diffuse transmittance from one side and the diffuse reflectance from the other side. The blue lines correspond to the cases when the final states are on the Ge side and the orange lines correspond to the cases where the final states are on the Si side. The diffuse transmittance and reflectance for evaluating the similarity are from the data presented in Fig. 4(a).

Lastly, we want to discuss the diffuse phonon scattering's role in interfacial transport. In Fig. 6(a), we plot the total transmission function as a function of frequency with different numbers of mixing layers. And we find that the total transmission can either be enhanced or reduced compared with the perfect Si/Ge interface as a result of the competition between the specular transmission versus diffuse transmission. From Figs. 6(b) and 6(c), we find as the degree of disorder increases, the specular transmission decreases while the diffuse transmission increases. In other words, the disorders remove specular channels while creating new diffuse channels. As a result, we do not observe significant changes in the transmission function due to atomic mixing. The opposite trends for specular and diffuse transmission versus the degree of disorder eventually leads to the maximum thermal conductance for 4 ml structures. It is interesting to note that similar enhancements for transmission enabled by disorders have been discovered in electron transport in heterostructures [38]. From Figs. 6(d)–6(i), we learn that the increasing amount of disorders always reduce the specular reflection function and increases the diffuse reflection function. Thus, the disorders can both increase the diffuse transmission (enhancing the interface conductance) and the diffuse reflection (worsening the interface conductance). This competition is another reason why we cannot observe significant enhancement of thermal conductance by disorders. Our current analysis is based on elastic scattering, yet phonon anharmonicity can contribute to interfacial transport by enabling vertical coupling between conduction channels of different frequencies. There are several works on understanding the anharmonicity's role in thermal interface conductance for abrupt interfaces [39–42], but how anharmonicity affects heat conduction channels through disordered interfaces remains unclear.

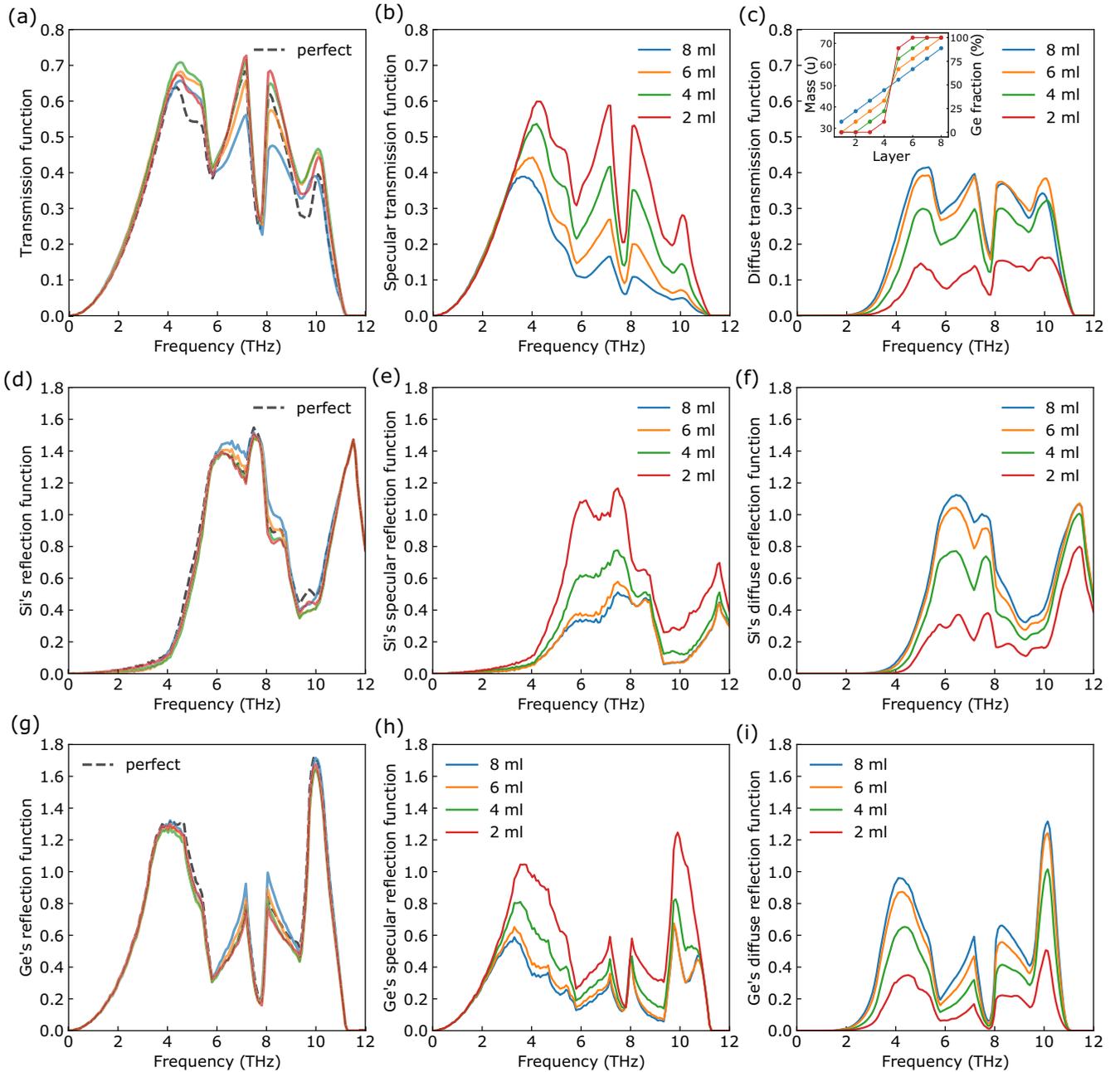


FIG. 6. (a) The total transmission function, (b) the specular transmission function, and (c) the diffuse transmission function versus phonon frequency for [001] Si/Ge interfaces from AGF. Inset: The ensemble-averaged mass profile as a function of atom layer number at the interface region obtained by averaging the mass of atoms within the same layer. The right axis is the Ge fraction in each atom layer. The distance between adjacent atom layers is $a/4 = 1.382 \text{ \AA}$. (d)–(f) The total, specular and diffuse reflection function from the Si side. (g)–(i) The total, specular, and diffuse reflection function from the Ge side.

IV. CONCLUSION

Through AGF calculation, we have demonstrated that the diffuse phonon scattering by a single disordered interface depends both on initial incoming states as well as the final outgoing states. The transmittance and reflectance strongly depend on the polar angle of group velocity. Also, the transmittance from one side and reflectance from the other side are generally different. That is to say, phonons do not lose their memory after diffuse scattering by a single interface. When two materials are similar, the diffuse transmittance from one

side and the diffuse reflectance from the other side become similar to each other. However, the total transmittance from one side and total reflectance from the other side are still different.

The number of specular transmission channels for interfacial transport is always reduced by interfacial disorders while new transmission channels are created by diffuse phonon scatterings. The competing roles of specular and diffuse transmission can lead to either enhanced or reduced transmission function and interfacial thermal conductance.

We also derived the expressions for transmittance and reflectance for diffuse scattering processes based on the continuum approximation, which works reasonably well in the low-frequency range. The model leads to different analytical expressions for diffuse transmittance from one side and diffuse reflectance from the other side. Our model also shows that the diffuse transmission opens up new transmission channels even for those states above the critical angle for total internal reflection.

ACKNOWLEDGMENTS

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APPENDIX: THE CONTINUUM MODELING OF DIFFUSE PHONON SCATTERING

1. The transmission and reflection matrix

We consider interfacial transport in the case of scalar phonon model in the continuum limit, where the equation of motion (EOM) for displacements writes [43]

$$\rho(\mathbf{r}) \frac{\partial^2 u}{\partial t^2} - \nabla \cdot (\mu(z) \nabla u) = 0, \quad (\text{A1})$$

where $\mu(z)$ is the bulk modulus. For an interface between two dissimilar solids, we have $\mu(z) = \mu_L$ for $z < 0$ and $\mu(z) = \mu_R$ for $z \geq 0$. $\rho(\mathbf{r}) = \rho_0(z) + \Delta\rho(\mathbf{r})$ is the density, where $\rho_0(z)$ is the density without mass disorder at the interface, expressed by $\rho_0(z < 0) = \rho_L$ and $\rho_0(z > 0) = \rho_R$. $\Delta\rho(\mathbf{r}) = \sum_i \Delta m_i \delta(\mathbf{r}_{\parallel} - \mathbf{r}_{i,\parallel}) \delta(z)$ is the density fluctuations due to atomic mixing at the interface, where Δm_i is the change of mass at atom site i and $\mathbf{r}_{i,\parallel}$ is the in-plane position of atom site i . The choice of the delta function form for density fluctuations suggests that the atomic mixing only exists exactly at the interface, thus our model does not apply to the cases where the atomic mixing exists even far away from the interface. When atomic mixing is realized by randomly swapping pairs of atoms on two sides of the interface, the average of mass fluctuations is zero $\sum_i \Delta m_i = 0$. The mass disorders are distributed randomly in the x - y plane. The ensemble average of any physical quantity P over many configurations of mass disorders is obtained by integrating over all possible positions of mass disorders,

$$\langle P \rangle = \int \prod_j \frac{d^2 \mathbf{r}_{\parallel,j}}{A} P, \quad (\text{A2})$$

where A is the cross-section area. After ensemble average, the average of mass fluctuations is still zero ($\sum_i \Delta m_i = 0$). We further assume an independent distribution of mass fluctuations such that

$$\left\langle \sum_{i,j} \Delta m_i \Delta m_j \right\rangle = \sum_i \langle (\Delta m_i)^2 \rangle. \quad (\text{A3})$$

The time-harmonic solution of Eq. (A1) reads

$$u = \sum_{\mathbf{q}_{\parallel}} u_{\mathbf{q}_{\parallel}}(z) \frac{e^{i(-\omega t + \mathbf{q}_{\parallel} \cdot \mathbf{r}_{\parallel})}}{\sqrt{A}}, \quad (\text{A4})$$

where $\mathbf{q}_{\parallel} = (q_x, q_y)$ is the transverse wave vector, ω is the phonon frequency, $\mathbf{r}_{\parallel} = (x, y)$ is the transverse position and $u_{\mathbf{q}_{\parallel}}(z)$ is the z -dependent component of the solution. The perpendicular wave vectors $q_{\perp}(z) = q_{z,L}$, when $z < 0$, and $q_{\perp}(z) = q_{z,R}$, when $z \geq 0$, are determined by the dispersion relation $\omega^2 = c_{L/R}^2 (q_{\parallel}^2 + q_{\perp,L/R}^2)$. Here, $c_{L/R}$ is the sound velocity defined by $c_{L/R} = \sqrt{\mu_{L/R}/\rho_{L/R}}$. Plugging in the time-harmonic solution to EOM, we have the following equation for $u_{\mathbf{q}_{\parallel}}(z)$:

$$\sum_{\mathbf{q}_{\parallel}} [(\rho_0(z) + \Delta\rho(\mathbf{r}))\omega^2 + \nabla \cdot \mu(z) \nabla] u_{\mathbf{q}_{\parallel}}(z) \frac{e^{i\mathbf{q}_{\parallel} \cdot \mathbf{r}_{\parallel}}}{\sqrt{A}} = 0. \quad (\text{A5})$$

Multiply both sides of Eq. (A5) by $\int d^2 \mathbf{r}_{\parallel} e^{-i\mathbf{q}'_{\parallel} \cdot \mathbf{r}_{\parallel}} / \sqrt{A}$. The orthogonality relations for plane waves leads to

$$\left[\mu(z) q_{\perp}^2(z) + \frac{\partial}{\partial z} \mu(z) \frac{\partial}{\partial z} \right] u_{\mathbf{q}'_{\parallel}} = \sum_{\mathbf{q}_{\parallel}} M_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}} \delta(z) u_{\mathbf{q}_{\parallel}}. \quad (\text{A6})$$

The scattering matrix $M_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}}$ is defined by

$$M_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}} = - \sum_i \Delta m_i \omega^2 A^{-1} e^{i(\mathbf{q}_{\parallel} - \mathbf{q}'_{\parallel}) \cdot \mathbf{r}_{i,\parallel}}. \quad (\text{A7})$$

The solution to Eq. (A6) is expressed by

$$u_{\mathbf{q}_{\parallel}}(z) = \delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} e^{iq_L z} + r_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \frac{\sqrt{v'_L}}{\sqrt{v_L}} e^{-iq_L z}, \quad z \leq 0, \\ u_{\mathbf{q}_{\parallel}}(z) = t_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \frac{\sqrt{\rho_L v'_L}}{\sqrt{\rho_R v_R}} e^{iq_R z}, \quad z \geq 0, \quad (\text{A8})$$

where $q_{L/R} = q_{z,L/R}$ is the phonon wave vector normal to the interface. $t_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}$ is the transmission matrix, and $r_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}$ is the reflection matrix. $v_{L/R} = c_{L/R} \cos \theta_{L/R}$ is the group velocity perpendicular to the interface, where $\theta_{L/R}$ is the angle between the direction of phonon velocity and the axis normal to the interface.

The transmission and reflection probability matrix are defined by the ratio of the transmitted flux normal to the interface of phonon \mathbf{q}_{\parallel} and the reflected flux normal to the interface of phonon \mathbf{q}_{\parallel} to the incident flux normal to the interface of phonon state \mathbf{q}'_{\parallel} , $T_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = \frac{J_{t, \mathbf{q}_{\parallel}}}{J_{\text{inc}, \mathbf{q}'_{\parallel}}}$ and $R_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = \frac{J_{r, \mathbf{q}_{\parallel}}}{J_{\text{inc}, \mathbf{q}'_{\parallel}}}$, respectively, where the time-averaged energy flux for a phonon mode reads [44]

$$J = \frac{\mu(z)}{-i\omega A} \int \left(u^* \frac{\partial u}{\partial z} - u \frac{\partial u^*}{\partial z} \right) d^2 \mathbf{r}_{\parallel}. \quad (\text{A9})$$

The resultant expressions for transmission probability matrix and reflection probability matrix are

$$T_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = |t_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}|^2, \quad (\text{A10a})$$

$$R_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = |r_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}|^2. \quad (\text{A10b})$$

The boundary conditions for displacement $u_{\mathbf{q}_{\parallel}}(z)$ for solving the transmission and reflection matrix write

$$u_{\mathbf{q}_{\parallel}}(0^-) = u_{\mathbf{q}_{\parallel}}(0^+), \quad (\text{A11a})$$

$$\mu(z) \frac{\partial}{\partial z} u_{\mathbf{q}_{\parallel}}(z) \Big|_{0^-}^{0^+} = \sum_{\mathbf{q}'_{\parallel}} M_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} u_{\mathbf{q}'_{\parallel}}(0), \quad (\text{A11b})$$

where the second boundary condition is obtained by integrating Eq. (A6) from $-\eta$ to η , with $\eta \rightarrow 0^+$. Plug in the expression in Eq. (A8) into the boundary condition. We obtain the following expressions:

$$\delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} + r_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = t_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}, \quad (\text{A12a})$$

$$\sum_{\mathbf{q}'_{\parallel}} (\delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} + i\Gamma_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}) t_{\mathbf{q}'_{\parallel}, \mathbf{q}'_{\parallel}} = \Lambda_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}, \quad (\text{A12b})$$

where

$$\Gamma_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = \frac{M_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}}{2\omega\bar{\rho}\bar{v}} \sqrt{\frac{v_R}{v_R''}}, \quad (\text{A13a})$$

$$\Lambda_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} = \delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \frac{\sqrt{v_L v_R}}{\bar{v}}, \quad (\text{A13b})$$

$$\bar{v} = \frac{\rho_L v_L + \rho_R v_R}{2\bar{\rho}}, \quad (\text{A13c})$$

$$\bar{\rho} = \sqrt{\rho_L \rho_R}. \quad (\text{A13d})$$

In particular, we can reorganize Eq. (A12b) and identify that the transmission matrix can be expanded in series as

$$t = \sum_{N=0}^{\infty} (-i\Gamma)^N \Lambda, \quad (\text{A14})$$

which is a summation of terms arising from multiple scatterings of different orders. We can discard high-order terms to obtain the approximate expression for the transmission matrix.

2. The Green's function in the continuum limit

The transmission matrix can be computed from the Green's function of the whole system. We choose to compute the Green's function because of the mathematical convenience in perturbation expansions using Dyson's equation. In the following, we will illustrate the exact relationship between the transmission matrix and the Green's function.

To start, we evaluate the unperturbed Green's function for a disorder-free interface. The unperturbed EOM writes

$$\left[\rho_0(z)\omega^2 + \frac{\partial}{\partial z} \mu(z) \frac{\partial}{\partial z} \right] u(z) = 0, \quad (\text{A15})$$

which can be identified as a Sturm-Liouville equation. Two sets of solutions are given by

$$u_{<}(z) = \begin{cases} t_L e^{-iq_L z}, & z < 0 \\ e^{-iq_R z} + r_L e^{iq_R z}, & z > 0 \end{cases} \quad (\text{A16})$$

and

$$u_{>}(z) = \begin{cases} t_R e^{iq_R z}, & z > 0 \\ e^{iq_L z} + r_R e^{-iq_L z}, & z < 0. \end{cases} \quad (\text{A17})$$

The continuity condition at interface gives

$$t_L = 1 + r_L = \frac{2\mu_R q_R}{\mu_L q_L + \mu_R q_R} \quad (\text{A18})$$

and

$$t_R = 1 + r_R = \frac{2\mu_L q_L}{\mu_L q_L + \mu_R q_R}. \quad (\text{A19})$$

For a Sturm-Liouville equation, the Wronskian writes [45]

$$W = u_{<}(z) \frac{du_{>}(z)}{z} - u_{>}(z) \frac{du_{<}(z)}{z} = \begin{cases} \frac{4i\mu_L q_L q_R}{\mu_L q_L + \mu_R q_R}, & z > 0 \\ \frac{4i\mu_R q_L q_R}{\mu_L q_L + \mu_R q_R}, & z < 0, \end{cases} \quad (\text{A20})$$

and the Green's function is defined by

$$G_0(z, z') = \begin{cases} \frac{u_{<}(z)u_{>}(z')}{\mu(z')W(z')}, & -\infty < z < z' \\ \frac{u_{>}(z')u_{<}(z)}{\mu(z)W(z)}, & z' < z < \infty, \end{cases} = \begin{cases} -\frac{i}{2\mu_L} \frac{t_R}{q_L} e^{-iq_L z} e^{iq_R z'}, & z < 0, z' > 0 \\ -\frac{i}{2\mu_R} \frac{t_L}{q_R} e^{-iq_L z'} e^{iq_R z}, & z > 0, z' < 0 \\ -\frac{i}{2\mu_L} \frac{e^{-iq_L|z'-z|+r_R e^{-iq_L(z+z')}}}{q_L}, & z < 0, z' < 0 \\ -\frac{i}{2\mu_R} \frac{e^{iq_R|z'-z|+r_L e^{iq_R(z+z')}}}{q_R}, & z > 0, z' > 0. \end{cases} \quad (\text{A21})$$

When z and z' both approach zero, the unperturbed Green's function at interface is

$$G_0^+ = -\frac{i}{\mu_L q_L + \mu_R q_R} = -\frac{i}{2\omega\bar{\rho}\bar{v}}, \quad (\text{A22})$$

where the superscript + is added to represent the retarded Green's function.

Then, we study the Green's function for the scenario where atomic mixing is present at the interface. It is convenient to define the Green's function operator,

$$\hat{G}^{\pm} = [\rho(\mathbf{r})\omega^2 - \hat{K} \pm i\eta]^{-1}, \quad (\text{A23})$$

where the operator $\hat{K} = -\nabla \cdot \mu(z)\nabla$ and η is an infinitesimal positive real number. The Green's function in the real space representation can then be expressed by $G^{\pm}(\mathbf{r}, \mathbf{r}') = \langle \mathbf{r} | \hat{G}^{\pm} | \mathbf{r}' \rangle$. The Green's function that describes the scattering channel between mode a of left side and mode b of right side is

$$G_{b,a}^{\pm}(z, z') = A^{-1} \iint d\mathbf{r}_{\parallel} d\mathbf{r}'_{\parallel} e^{-i(\mathbf{q}_{\parallel}\mathbf{r}_{\parallel} - \mathbf{q}'_{\parallel}\mathbf{r}'_{\parallel})} \langle u_{\mathbf{q}_{\parallel}} | \hat{G}^{\pm} | u'_{\mathbf{q}'_{\parallel}} \rangle, \quad (\text{A24})$$

where the transverse wave vector for mode a and mode b are \mathbf{q}'_{\parallel} and \mathbf{q}_{\parallel} , respectively.

When disorders are introduced, the perturbed eigenvector is related to the unperturbed eigenvector via [46]

$$u = u' + \int G^+(\mathbf{r}, \mathbf{r}') V(\mathbf{r}') u'(\mathbf{r}') d\mathbf{r}', \quad (\text{A25})$$

where u' is the eigenstate for the disorder-free case and the perturbation $V(\mathbf{r}) = -\Delta\rho(\mathbf{r})\omega^2$. Specifically, the second term

on the right-hand side of Eq. (A25) equals

$$\begin{aligned} & \int d\mathbf{r}' G^+(\mathbf{r}, \mathbf{r}') V(\mathbf{r}') u'(\mathbf{r}') \\ &= \int d\mathbf{r}' \{G^+(\mathbf{r}, \mathbf{r}') \hat{K}' - \delta(\mathbf{r} - \mathbf{r}') - [\hat{K}' G^+(\mathbf{r}, \mathbf{r}')]\} u'(\mathbf{r}'). \end{aligned} \quad (\text{A26})$$

Thus, Eq. (A25) is equivalent to

$$u = \int \mu(\mathbf{r}') \left(u' \frac{\partial G^+}{\partial z'} - G^+ \frac{\partial u'}{\partial z'} \right) \hat{\mathbf{e}}_z \cdot d\mathbf{S}'. \quad (\text{A27})$$

It is easy to show that

$$\begin{aligned} \langle u' | V | u \rangle_{b,a} &= \langle \hat{K}' u' | u \rangle_{b,a} - \langle u' | \hat{K}' | u \rangle_{b,a} \\ &= \int [u_a \hat{K}' u_b^* - u_b^* \hat{K}' u_a] d^3\mathbf{r} \\ &= \int \mu(\mathbf{r}) \left[u_b^* \frac{\partial u_a}{\partial z} - \frac{\partial u_b^*}{\partial z} u_a \right] \hat{\mathbf{e}}_z \cdot d\mathbf{S}, \end{aligned} \quad (\text{A28})$$

where we applied integration by part and the divergence theorem.

Plugging in the expression of u given by Eq. (A27) into Eq. (A28), we have

$$\begin{aligned} \langle u' | V | u \rangle_{b,a} &= \int [\hat{K}' u_b^*(\mathbf{r})] u_a(\mathbf{r}) d\mathbf{r} \\ &\quad - \iint u_b^*(\mathbf{r}) \mu(\mathbf{r}') \\ &\quad \times \left[u'_a(\mathbf{r}') \frac{\partial \hat{K}' G^+}{\partial z'} - \frac{\partial u'_a(\mathbf{r}')}{\partial z'} \hat{K}' G^+ \right] dS' d\mathbf{r} \\ &= \iint \mu(\mathbf{r}) \mu(\mathbf{r}') \left\{ \frac{\partial u_b^*(\mathbf{r})}{\partial z} \right. \\ &\quad \times \left[-u'_a(\mathbf{r}') \frac{\partial G^+}{\partial z'} + G^+ \frac{\partial u'_a(\mathbf{r}')}{\partial z'} \right] \\ &\quad \left. + u_b^*(\mathbf{r}) \left[u'_a(\mathbf{r}') \frac{\partial^2 G^+}{\partial z \partial z'} - \frac{\partial u'_a(\mathbf{r}')}{\partial z'} \frac{\partial G^+}{\partial z} \right] \right\} dS dS' \\ &= -4\mu_L \mu_{R,b}^* r_{b,R}^* e^{-ik_{b,R}z_2 + ik_{a,L}z'_1} G^+(z_2, z'_1) q_{b,R} q_{a,L} \\ &\quad - 4\mu_L^2 r_{b,R}^* e^{ik_{b,L}z_1 + ik_{a,L}z'_1} G_1^+(z_1, z'_1) q_{b,L} q_{a,L} \\ &\quad + 4\mu_L^2 r_{b,R}^* e^{ik_{b,L}z_1 - ik_{a,L}z'_1} G_2^+(z_1, z'_1) q_{b,L} q_{a,L}, \end{aligned} \quad (\text{A29})$$

where $G^+(z_1, z'_1) = G_1^+(z_1, z'_1) + G_2^+(z_1, z'_1)$ and we do not need to know the exact expression of G_1^+ and G_2^+ . Note that we have used the form of Green's function in its asymptotic limit in deriving the above expression. Denote L the length of the domain containing disorders. Then, $z_1, z'_1 < 0$ and $z_2, z'_2 > L$ are the boundary for integration. Since the random mass disorders are localized at the interface at $z = 0$, we have $L \rightarrow 0$, such that we can set $z_1 = z'_1 = 0^-$ and $z_2 = z'_2 = 0^+$.

Directly plugging in the general expression of u' for unperturbed system given by Eq. (A17) and u for perturbed system given by Eq. (A8), we can obtain another expression

for matrix element $\langle u' | V | u \rangle_{b,a}$,

$$\begin{aligned} \langle u' | V | u \rangle_{b,a} &= 2i\omega t_{b,R}^* r_{b,a} \sqrt{\rho_L v_{a,L} \rho_R v_{b,R}} \\ &\quad + 2i\omega \left(r_{b,R}^* r_{b,a} \frac{\sqrt{v_{a,L}}}{\sqrt{v_{b,L}}} - \delta_{b,a} \right) \rho_L v_{b,L}. \end{aligned} \quad (\text{A30})$$

By equating Eq. (A29) to Eq. (A30), we have found the relationship between transmission matrix and Green's function:

$$\begin{aligned} t_{b,a} &= 2i\omega G_{b,a}^+(0^+, 0^-) \sqrt{\rho_L \rho_R v_{a,L} v_{b,R}} \\ &= 2i\omega \bar{\rho} \sqrt{v_R v_L} G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^+. \end{aligned} \quad (\text{A31})$$

Furthermore, using the boundary condition given by Eq. (A12a), we find that the reflection matrix is related to Green's function through

$$\begin{aligned} r_{b,a} &= 2i\omega G_{b,a}^+(0^-, 0^-) \rho_L \sqrt{v_{a,L} v_{b,L}} - \delta_{ba} \\ &= 2i\omega \rho_L \sqrt{v_L v_L} G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^+ - \delta_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}. \end{aligned} \quad (\text{A32})$$

3. The ensemble averaged Green's function

From the series expansion of the transmission matrix in Eq. (A14) and relationship between transmission matrix and Green's function given by Eq. (A31), we can obtain the following series for the ensemble averaged Green's function:

$$\langle G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^+ \rangle = -i \frac{1}{2\omega \bar{\rho} \bar{v}'} \frac{\sqrt{v_R}}{\sqrt{v_R}} \sum_N \langle (-i\Gamma)^N \rangle_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}. \quad (\text{A33})$$

According to Eq. (A2), the ensemble average of matrix $(-i\Gamma)^N$ is obtained by integrating over all possible impurity positions:

$$\langle (-i\Gamma)^N \rangle = \int \prod_j \frac{d^2 \mathbf{r}_{\parallel, j}}{A} (-i\Gamma)^N. \quad (\text{A34})$$

In the weak perturbation limit, using Eqs. (A3), (A7), and (A13a), we write down the Green's function in Eq. (A33) up to the second order,

$$\langle G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^+ \rangle = \left(G_0^+(\mathbf{q}'_\parallel) + G_0^{+2}(\mathbf{q}'_\parallel) \frac{V_2}{A} \sum_{\mathbf{q}''_\parallel} G_0^+(\mathbf{q}''_\parallel) \right) \delta_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}, \quad (\text{A35})$$

where $V_2 = \langle \sum_i \frac{m_i^2}{A} \omega^4 \rangle$. The first-order term vanishes due to ensemble average $\langle \sum_i m_i \omega^2 \rangle = 0$. The diagonal form of Eq. (A35) implies that the ensemble average recovers the in-plane translational symmetry of the unperturbed Green's function.

4. The transmission and reflection probability matrix

From Eq. (A10a), we find that the transmission probability matrix is related to the product of retarded and advanced Green's function:

$$\langle T_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel} \rangle = 4\omega^2 \bar{\rho}^2 v'_L v_R \langle G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^+ G_{\mathbf{q}_\parallel, \mathbf{q}'_\parallel}^- \rangle. \quad (\text{A36})$$

The ensemble averaged $G_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}^+ G_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}^-$ can be expressed by

$$\begin{aligned} \langle G_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}^+ G_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}^- \rangle &= |\langle G_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}}^+ \rangle|^2 + \sum_{\mathbf{q}''_{\parallel}, \mathbf{q}'''_{\parallel}} |\langle G^+(\mathbf{q}_{\parallel}, \mathbf{q}''_{\parallel}) \rangle|^2 W_{\mathbf{q}''_{\parallel}, \mathbf{q}'''_{\parallel}} |\langle G^+(\mathbf{q}''_{\parallel}, \mathbf{q}'_{\parallel}) \rangle|^2, \end{aligned} \quad (\text{A37})$$

where the term $W_{\mathbf{q}''_{\parallel}, \mathbf{q}'''_{\parallel}}$ is called the reducible vertex function. To the lowest order, the vertex function reads [47]

$$W_{\mathbf{q}''_{\parallel}, \mathbf{q}'''_{\parallel}} = \frac{V_2}{A}. \quad (\text{A38})$$

From Eq. (A35) to Eq. (A38), we obtain the expression for the transmission probability matrix,

$$\begin{aligned} \langle T_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \rangle &= \delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \frac{4\rho_L \rho_R v_L v_R}{|\rho_L v_L + \rho_R v_R|^2} \\ &\times \left[1 - 2\text{Re} \mathcal{G}^+ \omega^{-1} V_2 \left(\frac{1}{\rho_L v_L + \rho_R v_R} \right) \right] \\ &+ \frac{4\omega^{-2} V_2}{A} \frac{\rho_R v_R}{|\rho_L v_L + \rho_R v_R|^2} \frac{\rho_L v'_L}{|\rho_L v'_L + \rho_R v'_R|^2}, \end{aligned} \quad (\text{A39})$$

where $\mathcal{G}^+ = (\sum_{\mathbf{q}''_{\parallel}} iG_0^+(\mathbf{q}''_{\parallel})) / A$ and the analytical expression for \mathcal{G}^+ can be found in the next session. We identify that the diagonal term is responsible for specular transmission, while the off-diagonal term is responsible for diffuse transmission. Similarly, from Eqs. (A10b), (A32), and (A35), we derive that the reflection probability matrix writes

$$\begin{aligned} \langle R_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \rangle &= \delta_{\mathbf{q}_{\parallel}, \mathbf{q}'_{\parallel}} \frac{|\rho_L v_L - \rho_R v_R|^2}{|\rho_L v_L + \rho_R v_R|^2} \\ &\times \left[1 - 4\text{Re} \mathcal{G}^+ \omega^{-1} V_2 \text{Re} \left(\frac{\rho_L v_L}{\rho_L^2 v_L^2 - \rho_R^2 v_R^2} \right) \right] \\ &+ \frac{4\omega^{-2} V_2}{A} \frac{\rho_L v_L}{|\rho_L v_L + \rho_R v_R|^2} \frac{\rho_L v'_L}{|\rho_L v'_L + \rho_R v'_R|^2}. \end{aligned} \quad (\text{A40})$$

5. The analytical expression of \mathcal{G}^+

The term \mathcal{G}^+ in Eqs. (A39) and (A40) can be written in terms of an integral over all transverse wave vectors,

$$\mathcal{G}^+ = \int \frac{d^2 \mathbf{q}_{\parallel}}{(2\pi)^2} \frac{1}{\sqrt{\mu_L(\rho_L \omega^2 - \mu_L q_{\parallel}^2)} + \sqrt{\mu_R(\rho_R \omega^2 - \mu_R q_{\parallel}^2)}}. \quad (\text{A41})$$

Introduce the ratio of bulk densities and moduli as $a = \rho_R / \rho_L$ and $b = \mu_R / \mu_L$. Depending on the bulk moduli and densities of two sides, the expression for the real part of \mathcal{G}^+ is as follows. When $(b - a)(1 - b^2) > 0$,

$$\begin{aligned} \text{Re} \mathcal{G}^+ &= \frac{\omega \mathcal{G}_0^+}{1 - b^2} \left[1 - \sqrt{ab} + \sqrt{\frac{b(b-a)}{1-b^2}} \right] \\ &\times \left(\text{atan} \sqrt{\frac{a(1-b^2)}{b-a}} - \text{atan} \sqrt{\frac{1-b^2}{b(b-a)}} \right), \end{aligned} \quad (\text{A42})$$

where $\mathcal{G}_0^+ = \frac{1}{2\pi\mu_L} \sqrt{\frac{\rho_L}{\mu_L}}$ and for Si, $\mathcal{G}_0^+ = 2.62 \times 10^{-16} \text{s}^3/\text{kg}$. When $(b - a)(1 - b^2) < 0$,

$$\begin{aligned} \text{Re} \mathcal{G}^+ &= \frac{\omega \mathcal{G}_0^+}{1 - b^2} \left[1 - \sqrt{ab} + \frac{1}{2} \sqrt{\frac{b(a-b)}{1-b^2}} \right] \\ &\times \left(\ln \frac{|1 - \sqrt{\frac{b(a-b)}{1-b^2}}|}{1 + \sqrt{\frac{b(a-b)}{1-b^2}}} - \ln \frac{|1 - \sqrt{\frac{a-b}{a(1-b^2)}}|}{1 + \sqrt{\frac{a-b}{a(1-b^2)}}} \right). \end{aligned} \quad (\text{A43})$$

When $b = 1$ and $a \neq 1$,

$$\text{Re} \mathcal{G}^+ = \frac{\omega \mathcal{G}_0^+}{3} \frac{1 - a^{\frac{3}{2}}}{1 - a}. \quad (\text{A44})$$

When $a = b$,

$$\text{Re} \mathcal{G}^+ = \frac{\omega \mathcal{G}_0^+}{1 + b}. \quad (\text{A45})$$

6. The specular and diffuse transmittance/reflectance

The transmittance for a given initial state \mathbf{q}_{\parallel} is defined by summing transition probabilities to different final states \mathbf{q}'_{\parallel} ,

$$T_{L \rightarrow R}(\mathbf{q}_{\parallel}) = \sum_{\mathbf{q}'_{\parallel}} \langle T_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}} \rangle, \quad (\text{A46})$$

where $\langle T_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}} \rangle$ is the transmission probability matrix defined in Eq. (A39).

In the following, we will use direction $\Omega_L = (\theta_L, \phi_L)$ to denote a phonon state \mathbf{q}_{\parallel} , where $(\mathbf{q}_{\parallel}, q_L) = \frac{\omega}{c_L} (\sin\theta_L \cos\phi, \sin\theta_L \sin\phi, \cos\theta_L)$. Note that the group velocity is parallel to the wave vector thus the angles for the group velocity and the wave vector are the same. After integration, the transmittance in Eq. (A46) is given by

$$\begin{aligned} T_{L \rightarrow R}(\Omega_L) &= T_{s,L \rightarrow R}(\Omega_L) + T_{d,L \rightarrow R}(\Omega_L) \\ &= T_{\text{AMM}}(\Omega_L) p_T(\Omega_L) + T_{d,L \rightarrow R}(\Omega_L). \end{aligned} \quad (\text{A47})$$

The first term is the specular transmittance, which is the product of transmittance from AMM [2,4],

$$T_{\text{AMM}}(\Omega_L) = \frac{4\rho_L \rho_R v_L v_R}{|\rho_L v_L + \rho_R v_R|^2}, \quad (\text{A48})$$

and the specularly parameter for transmittance:

$$p_T(\Omega_L) = 1 - 2\text{Re} \mathcal{G}^+ \omega^{-1} V_2 \left(\frac{1}{\rho_L v_L + \rho_R v_R} \right). \quad (\text{A49})$$

The second term, diffuse transmittance, is given by Eq. (13) in Sec. II.

Similarly, the reflectance for a given incident state from the left side is given by

$$\begin{aligned} R_{L \rightarrow L}(\Omega_L) &= \sum_{\mathbf{q}'_{\parallel}} \langle R_{\mathbf{q}'_{\parallel}, \mathbf{q}_{\parallel}} \rangle \\ &= R_{s,L \rightarrow L}(\Omega_L) + R_{d,L \rightarrow L}(\Omega_L) \\ &= R_{\text{AMM}}(\Omega_L) p_R(\Omega_L) + R_{d,L \rightarrow L}(\Omega_L), \end{aligned} \quad (\text{A50})$$

where $\langle R_{\mathbf{q}_{\parallel}, \mathbf{q}_{\parallel}} \rangle$ is the reflection probability matrix defined in Eq. (A40). The reflectance by AMM writes

$$R_{\text{AMM}}(\Omega_L) = \frac{|\rho_L v_L - \rho_R v_R|^2}{|\rho_L v_L + \rho_R v_R|^2}. \quad (\text{A51})$$

The specularity parameter for reflectance is given by

$$p_R(\Omega_L) = 1 - 4\text{Re} \mathcal{G}^+ \omega^{-1} V_2 \text{Re} \left(\frac{\rho_L v_L}{\rho_L^2 v_L^2 - \rho_R^2 v_R^2} \right) \quad (\text{A52})$$

and the diffuse reflectance $R_{d,L \rightarrow L}(\Omega_L)$ is defined by Eq. (15). We want to stress that the expressions in Eqs. (A47) and (A50) add up to one in the current lowest-order perturbation theory, which means our continuum model is a self-consistent theory. However, this is not a guaranteed property at higher orders.

In the previous study of partially specular and partially specular interface scattering by a disordered interface [48], the transmittance and reflectance are often phenomenologically written as

$$T(\Omega) = p(\Omega)T_{\text{AMM}}(\Omega) + (1 - p(\Omega))T_{\text{DMM}}(\Omega), \quad (\text{A53a})$$

$$R(\Omega) = p(\Omega)R_{\text{AMM}}(\Omega) + (1 - p(\Omega))R_{\text{DMM}}(\Omega), \quad (\text{A53b})$$

where p is the specularity parameter calculated by Ziman's equation [49]. However, in our continuum modeling, there are two specularity parameters, one for transmittance [Eq. (A49)], one for reflectance [Eq. (A52)] and they are generally not equal. The necessity of two specularity parameters has been hypothesized by Li *et al.* [50] and our analytical model gives direct support for the hypothesis of two different specularity parameters. What's more, it is entirely possible to have p_R in our model larger than one (this is also observed in AGF calculation presented in the Supplemental Material [28]), while the specularity parameter p given by Ziman's equation is bounded by one. Thus, the specularity parameter is merely a correction factor and cannot be interpreted as probability of being specularly scattered.

Similar to Eq. (20), we further compute the frequency-resolved average transmittance by integrating over solid angle

$$T_{L \rightarrow R}(\omega) = 2 \int_0^{\pi/2} d\theta \sin\theta \cos\theta T_{L \rightarrow R}(\Omega_L), \quad (\text{A54})$$

and the reflectance can be similarly computed. The energy-resolved transmission function, which measures the number of conduction channels for interfacial thermal transport, is obtained by

$$\begin{aligned} \Theta(\omega) &= A \int \frac{d^2 \mathbf{q}_{\parallel}}{(2\pi)^2} T_{L \rightarrow R}(\omega, \mathbf{q}_{\parallel}) \\ &= 2\pi A \int \frac{d^2 \mathbf{q}_{\parallel} dq_L}{(2\pi)^3} T_{L \rightarrow R}(\Omega_L) v_L \delta(\omega - c_L \sqrt{q_{\parallel}^2 + q_L^2}) \\ &= \pi A D_L(\omega) c_L \int_0^{\pi/2} d\theta \sin\theta \cos\theta T_{L \rightarrow R}(\Omega_L), \end{aligned} \quad (\text{A55})$$

where $D_L(\omega) = \frac{\omega^2}{2\pi^2 c_L^3}$ is the density of states of left side and $\sin\theta = |\mathbf{q}_{\parallel}|c_L/\omega$. The two-probe interfacial thermal conduc-

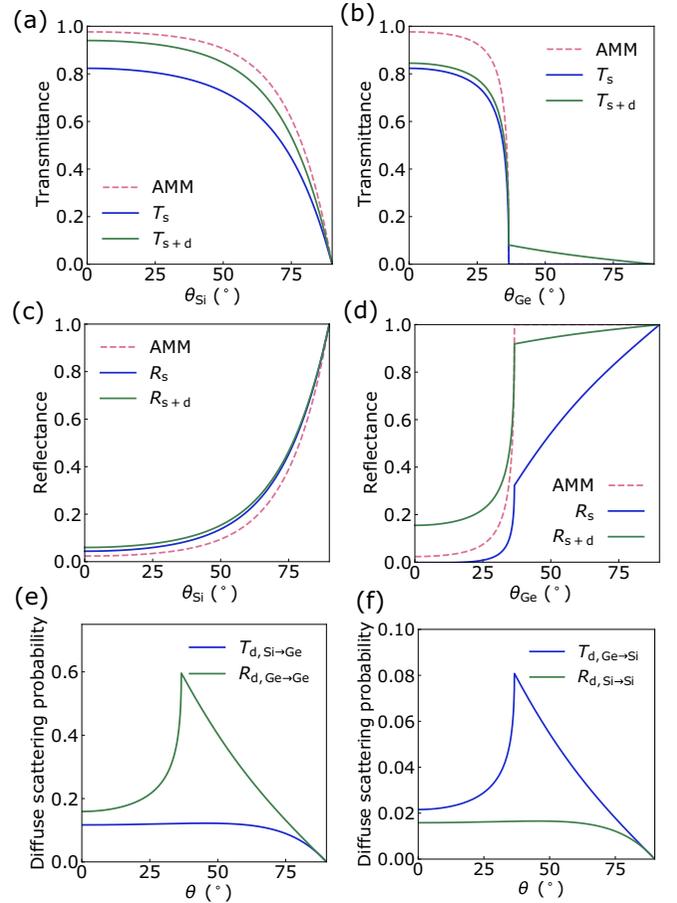


FIG. 7. The transmittance and reflectance for a rough Si/Ge interface predicted from the continuum model. (a), (b) The specular and diffuse transmittance of acoustic phonons at 4 THz in Si and Ge compared with AMM. (c), (d) The specular and diffuse reflectance of acoustic phonons at 4 THz in Si and Ge compared with AMM. (e), (f) The diffuse transmittance from one side and reflectance from the other side of acoustic phonons at 4 THz. θ is the velocity angle of the incident state.

tance per unit area is determined by the transmission function,

$$G = \frac{1}{2\pi A} \int_0^\infty \hbar\omega \Theta(\omega) \frac{\partial f(\omega, T)}{\partial T} d\omega, \quad (\text{A56})$$

where $f(\omega, T)$ is the Bose-Einstein distribution function.

7. The interface scattering transition probability for a rough Si/Ge interface

We apply the derived equations for transmittance and reflectance for a rough Si/Ge interface along [001] direction. If we assume the atomic mixing is realized by swapping Si and Ge atoms on two sides of interface, the variance of mass fluctuations is estimated to be $\langle m_i^2 \rangle \approx (m_{\text{Si}} - m_{\text{Ge}})^2 = 1.985 \times 10^3 \text{u}^2$. Thus, the parameter $V_2 = n \langle m_i^2 \rangle \omega^4$, where n is the number of pairs of swapped Si and Ge atoms per unit area. We choose $n = 2/a^2$ in the following calculation, and $a = 5.527 \text{ \AA}$ is the lattice constant, obtained by taking the average of Si's and Ge's lattice constants. The bulk moduli

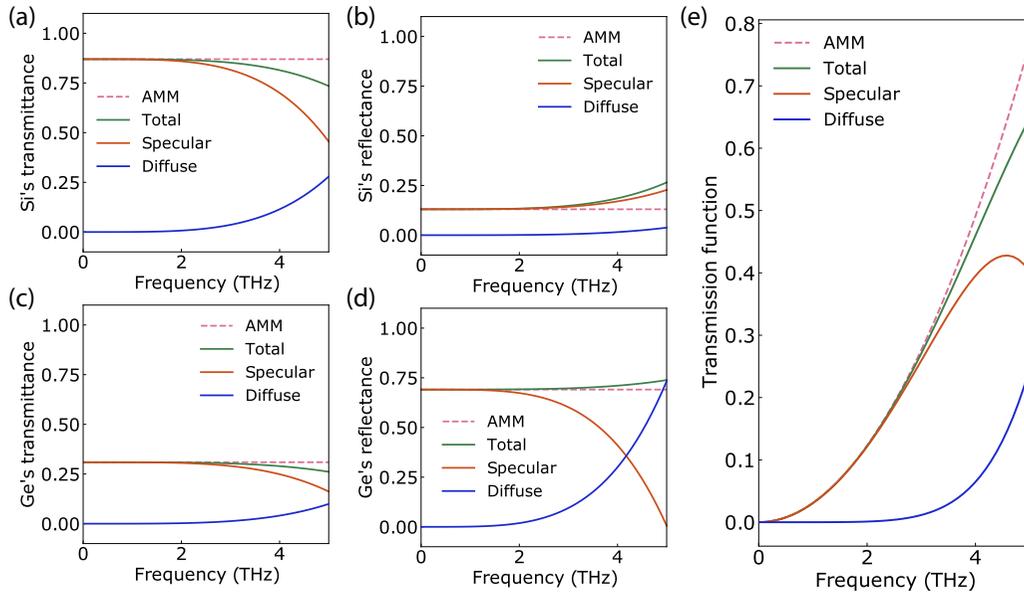


FIG. 8. (a)–(d) The frequency-resolved transmittance and reflectance from Si side and Ge side from continuum modeling. (e) The transmission function $\Theta(\omega)$ for Si/Ge interface as a function of frequency. A multiplicity factor of 3 is multiplied in the transmission function as there are three acoustic phonon branches. When the frequency is much higher than 5 THz, the lowest perturbation theory is no longer valid, as the perturbed part becomes large.

of Si and Ge are $\mu_L = 95$ GPa and $\mu_R = 77.2$ GPa. The densities of Si and Ge are $\rho_L = 2.329 \times 10^3$ kg/m³ and $\rho_R = 5.323 \times 10^3$ kg/m³.

As shown in Fig. 7, we find that the phonon transmittance of Si is smaller compared with AMM. Although there are more transmission channels due to diffuse scattering, the reduction of transmittance is mainly due to fewer specular transmission channels, which are removed by interfacial disorders. In addition, we note that the diffuse transmission from the Ge side opens new transmission channels above the critical angle for total reflection. Furthermore, we find that the reflectance from the Si side increases with the angle, similar to the trend of AMM. For reflectance from the Ge side, the specular part is smaller than AMM. Due to large diffuse reflectance shown in Fig. 7(e), the total reflection probability is eventually higher than predictions of AMM below the critical angle and lower above the critical angle. In fact, from the expression of Eqs. (A39) and (A40), we find that the specular transmittance is always reduced by disorders while the specular reflectance can either be enhanced or reduced depending on the sign of $\rho_L v_L - \rho_R v_R$.

From Figs. 7(e) and 7(f), we observe that generally the diffuse transmittance from one side is different from the diffuse reflectance from the other side. Furthermore, we find that as frequency increases, the specular scattering probability decreases while the diffuse scattering probability increases,

as shown in Figs. 8(a)–8(d). For the transmittance from both sides, the reduction in the specular part is always larger than the increment in the diffuse part, hence a reduced total transmittance. In contrast, for reflectance, the increment in the diffuse part prevails over the reduction in the specular part, causing a greater total reflectance. When we compare the phonon transmission function for the Si/Ge interface in Fig. 8(e), the interface disorders lead to a smaller total transmission, thus a smaller thermal conductance. Note that when $\omega > 5$ THz, the specular reflectance from Ge side will become negative, because the perturbation is no longer a small quantity. From Eq. (A52), we see that the reduction in the specularly parameter for reflectance p_R varies drastically with frequency with ω^4 scaling. When $p_R \sim 0$, we have a critical frequency $\omega \sim \left(\frac{\mu^2 A}{m_i^2}\right)^{1/4}$ and our model only works below this critical frequency.

To summarize, continuum modeling using perturbation theory to the lowest order suggests that the diffuse scattering cannot make a phonon forget its origin, opposing the picture of DMM. However, we want to point out limitations of the continuum model of scalar phonons. First, the phonon mode conversion is not considered. Second, the model is valid for low-frequency acoustic phonons thus at lifted temperatures, where high-frequency phonons are playing an important role in interfacial phonon transport, the model is no longer valid.

- [1] E. T. Swartz and R. O. Pohl, *Rev. Mod. Phys.* **61**, 605 (1989).
- [2] W. Little, *Can. J. Phys.* **37**, 334 (1959).
- [3] P. Kapitza, *J. Phys. USSR* **4**, 181 (1941).
- [4] I. M. Khalatnikov, *Zh. Eksp. Teor. Fiz.* **22**, 687 (1952).
- [5] H.-K. Lyo and D. G. Cahill, *Phys. Rev. B* **73**, 144301 (2006).

- [6] Z. Cheng, Y. R. Koh, H. Ahmad, R. Hu, J. Shi, M. E. Liao, Y. Wang, T. Bai, R. Li, E. Lee *et al.*, *Commun. Phys.* **3**, 115 (2020).
- [7] Y. Chalopin, K. Esfarjani, A. Henry, S. Volz, and G. Chen, *Phys. Rev. B* **85**, 195302 (2012).
- [8] K. Gordiz and A. Henry, *New J. Phys.* **17**, 103002 (2015).

- [9] K. Gordiz and A. Henry, *Sci. Rep.* **6**, 23139 (2016).
- [10] E. S. Landry and A. J. H. McGaughey, *Phys. Rev. B* **80**, 165304 (2009).
- [11] K. Sääskilähti, J. Oksanen, J. Tulkki, and S. Volz, *Phys. Rev. B* **90**, 134312 (2014).
- [12] N. Yang, T. Luo, K. Esfarjani, A. Henry, Z. Tian, J. Shiomi, Y. Chalopin, B. Li, and G. Chen, *J. Comput. Theor. Nanosci.* **12**, 168 (2015).
- [13] Y. Chalopin and S. Volz, *Appl. Phys. Lett.* **103**, 051602 (2013).
- [14] C. Kimmer, S. Aubry, A. Skye, and P. K. Schelling, *Phys. Rev. B* **75**, 144105 (2007).
- [15] B. Deng, A. Chernatynskiy, M. Khafizov, D. H. Hurley, and S. R. Phillpot, *J. Appl. Phys.* **115**, 084910 (2014).
- [16] W. Zhang, T. S. Fisher, and N. Mingo, *J. Heat Transfer* **129**, 483 (2006).
- [17] W. Zhang, T. Fisher, and N. Mingo, *Numer. Heat Transfer, Part B* **51**, 333 (2007).
- [18] Z. Tian, K. Esfarjani, and G. Chen, *Phys. Rev. B* **86**, 235304 (2012).
- [19] Z.-Y. Ong and G. Zhang, *Phys. Rev. B* **91**, 174302 (2015).
- [20] S. Sadasivam, U. V. Waghmare, and T. S. Fisher, *Phys. Rev. B* **96**, 174302 (2017).
- [21] B. Latour, N. Shulumba, and A. J. Minnich, *Phys. Rev. B* **96**, 104310 (2017).
- [22] Z.-Y. Ong, G. Schusteritsch, and C. J. Pickard, *Phys. Rev. B* **101**, 195410 (2020).
- [23] Z.-Y. Ong, *Europhys. Lett.* **133**, 66002 (2021).
- [24] D. A. Young and H. J. Maris, *Phys. Rev. B* **40**, 3685 (1989).
- [25] H. Zhao and J. Freund, *J. Appl. Phys.* **97**, 024903 (2005).
- [26] S. Lu and A. J. McGaughey, *J. Appl. Phys.* **121**, 115103 (2017).
- [27] P. A. Khomyakov, G. Brocks, V. Karpan, M. Zwierzycki, and P. J. Kelly, *Phys. Rev. B* **72**, 035450 (2005).
- [28] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.104.085310> for details of our implementation of the mode-resolved atomistic Green's function formalism.
- [29] M. L. Sancho, J. L. Sancho, J. L. Sancho, and J. Rubio, *J. Phys. F* **15**, 851 (1985).
- [30] C. H. Lewenkopf and E. R. Mucciolo, *J. Comput. Electron.* **12**, 203 (2013).
- [31] F. Sols, M. Macucci, U. Ravaioli, and K. Hess, *J. Appl. Phys.* **66**, 3892 (1989).
- [32] At a given transverse wave vector \mathbf{q}_{\parallel} , there are multiple phonon states with different q_z or degenerate states with the same q_z . Here, we use \mathbf{q}_{\parallel} to describe one of those states.
- [33] V. Popescu and A. Zunger, *Phys. Rev. Lett.* **104**, 236403 (2010).
- [34] C. Caroli, R. Combescot, P. Nozieres, and D. Saint-James, *J. Phys. C* **4**, 916 (1971).
- [35] K. Bi, J. Wang, Y. Wang, J. Sha, Z. Wang, M. Chen, and Y. Chen, *Phys. Lett. A* **376**, 2668 (2012).
- [36] One may be concerned about the contribution from specularly parameter to the diffuse transmittance and reflectance. We have discussed the role of specularly parameter in diffuse scattering in the Supplemental Material [28].
- [37] L. Yang, B. Latour, and A. J. Minnich, *Phys. Rev. B* **97**, 205306 (2018).
- [38] D. Vashae and A. Shakouri, *Phys. Rev. Lett.* **92**, 106103 (2004).
- [39] X. Wu and T. Luo, *J. Appl. Phys.* **115**, 014901 (2014).
- [40] T. Feng, Y. Zhong, J. Shi, and X. Ruan, *Phys. Rev. B* **99**, 045301 (2019).
- [41] J. Dai and Z. Tian, *Phys. Rev. B* **101**, 041301(R) (2020).
- [42] Y. Guo, Z. Zhang, M. Bescond, S. Xiong, M. Nomura, and S. Volz, *Phys. Rev. B* **103**, 174306 (2021).
- [43] G. Eason, *Acta Mech.* **7**, 137 (1969).
- [44] S. G. Johnson, Notes on the algebraic structure of wave equations (2007).
- [45] L. Copley, in *Mathematics for the Physical Sciences* (De Gruyter Open Poland, Sciendo Migration, 2014), pp. 384–417.
- [46] A. Messiah, *Quantum Mechanics: Volume II* (North-Holland Publishing Company, Amsterdam, 1962).
- [47] D. A. Stewart, W. H. Butler, X.-G. Zhang, and V. F. Los, *Phys. Rev. B* **68**, 014433 (2003).
- [48] G. Chen, *Phys. Rev. B* **57**, 14958 (1998).
- [49] J. M. Ziman, *Electrons and Phonons: The Theory of Transport Phenomena in Solids* (Oxford University Press, 2001).
- [50] D. Li and A. J. McGaughey, *Nanoscale Microscale Thermophys. Eng.* **19**, 166 (2015).