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Lattice thermal conductivity evaluated using elastic properties

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Lattice thermal conductivity is one of the most important thermoelectric parameters in determining the energy conversion efficiency of thermoelectric materials. However, the lattice thermal conductivity evaluation requires time-consuming first-principles (quasi)phonon calculations, which limits seeking high-performance thermoelectric materials through high-throughput computations. Here, we establish a methodology to determine the Debye temperature Θ, Grüneisen parameter γ, and lattice thermal conductivity κ using computationally feasible elastic properties (the bulk and shear moduli). For 39 compounds with three different prototypes (the cubic isotropic rocksalt and zinc blende, and the noncubic anisotropic wurtzite), the theoretically calculated Θ, γ, and κ are in reasonable agreement with those determined using (quasi)harmonic phonon calculations or experimental measurements. Our results show that the methodology is an efficient tool to predict the anharmonicity and the lattice thermal conductivity.

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I. INTRODUCTION

Thermoelectric materials play many promising roles in solving problems of energy and environment. They can directly convert heat into electricity, and the conversion efficiency is characterized by the thermoelectric figure of merit $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, and κ is the thermal conductivity. κ includes the electronic (κ_{ele}) and lattice vibration (the dominating part, κ_L) contributions. The lattice thermal conductivity κ_L is, in principle, independent of the rest of the parameters (S, σ, and κ_e) and can be easily tuned using different methodologies. One of the methodologies to obtain the low thermal conductivity is seeking materials with ordered crystal structures exhibiting strong lattice anharmonicity [1], such as $AgSbTe_2$ [2], Cu_3SbSe_3 [3,4], $Cu_{12}Sb_4Se_{13}$ [5], and $Cu_{12}Sb_4S_{13}$[6]. Recently, Zhao *et al.* [7] discovered that SnSe has a maximum ZT of ~2.6 at ~900 K. The authors suggested that the high thermoelectric figure of merit was attributed to the extremely low lattice thermal conductivity of the SnSe crystals due to strong anharmonicity. Thus, understanding and evaluating the lattice thermal conductivity are important to seeking high-performance thermoelectric materials.

The current lattice thermal conductivity evaluations involve the first-principles phonon calculations, such as the full iterative solution to the phonon Boltzmann transport equation [8] (ShengBTE [9]) or the first-principles Debye-Callaway model combination with the relaxation-time approximation [4,10,11]. In previous studies of Cu_3SbSe_3 [4] and SnSe [1,7,12], the authors suggested that the strong lattice anharmonicity results from weak interatomic interactions, i.e., the weak Sb-Se and Sn-Se bonds in Cu_3SbSe_3 and SnSe, respectively. The weak interatomic interactions usually indicate that the crystal structure can be easily deformed under mechanical stress, resulting in the soft lattice or weak elastic properties. Zeier *et al.* roughly suggested the slow sound speed reflecting the low thermal conductivity [13]. Clarke [14] has derived a minimum thermal conductivity formula as a function of Young's modulus: the low thermal conductivity benefits from the small Young's modulus. The author applied the formula to identify thermal barrier coating candidates for extremely high temperature applications. However, this formula is not suitable for understanding the lattice thermal conductivity at a specific temperature.

Since acoustic modes play an important role in the heat transfer in semiconductors [15], Slack [15,16] provided an approach to express the lattice thermal conductivity (κ_L, or κ for simplicity) at a temperature of the form

$$\kappa_L = A\frac{M_a\delta n^{1/3}\Theta^3}{\gamma^2 T}, \tag{1}$$

where M_a, δ^3, n, Θ, γ, and A are the average atomic mass, the volume per atom, the number of atoms in the primitive unit cell, the acoustic Debye temperature, the acoustic Grüneisen parameter, and $A = \frac{2.43\times10^{-8}}{1-\frac{0.514}{\gamma}+\frac{0.228}{\gamma^2}}$, respectively. The formula has been widely used in the lattice thermal conductivity evaluation [2,3,15,17,18]. The Debye temperatures and Grüneisen parameters of acoustic branches can be accurately determined using phonon dispersions either from lattice dynamic calculations or experimental measurements [15,17]. These acoustic parameters are denoted as Θ_ω and γ_ω. The Grüneisen parameter γ_ω represents the strength of lattice anharmonicity of a compound. It requires the quasiharmonic phonon calculations or the experimentally measured thermal expansion coefficient. Without these, Xiao *et al.* [19] recently used an efficient formula that is the function of the elastic property, the Poisson ratio ν or the sound velocity, to estimate the Grüneisen parameter, and it seems that the formula results in reasonable Grüneisen parameters for four compounds (PbS, PbSe, PbTe, and SnSe). However, we find that the formula is not suitable for many other compounds (see below). Thus, it

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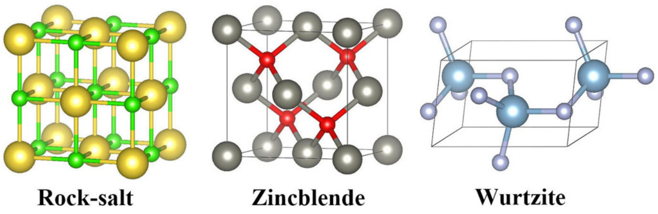

FIG. 1. Geometries of three prototype structures: rocksalt (RS), zinc blende (ZB), and wurtzite (WZ), from left to right.

is still a big challenge to efficiently evaluate the Grüneisen parameter (or the lattice anharmonicity) of a compound.

To avoid first-principles (quasi)harmonic phonon calculations, we develop a methodology to efficiently estimate the Grüneisen parameter and lattice thermal conductivity based on the first-principles-determined elastic properties (the speed of sound, bulk modulus, and shear modulus). For 39 compounds with three different prototype structures [rocksalt (RS), zinc blende (ZB), and wurtzite (WZ); Fig. 1], they represent a variable lattice thermal conductivity behavior, 0.4–350 W/mK at 300 K from the experimental measurements [17]. These well-studied data are good candidates to calibrate the results from developed methodologies, such as the theoretically calculated lattice thermal conductivity by solving the linearized phonon Boltzmann equation [20] or using the Debye-Callaway approach [17]. For the above compounds, our elastic-property-estimated Grüneisen parameter, Debye temperature, and lattice thermal conductivity are in reasonable agreement with those using the lattice dynamic calculations and the experimental measurements. Our work establishes a straightforward method to estimate the Grüneisen parameter (or anharmonicity) of a compound using computationally feasible elastic properties.

II. METHODOLOGY

We perform the first-principles calculations with density function theory using the Vienna Ab initio Simulation Package (VASP) [21]. The interactions between electrons are described by the projector augmented-wave (PAW) method [22]. The electronic exchange-correlation functional is accounted for by the generalized gradient approximation of Perdew, Burke, and Ernzerhof (GGA-PBE) [23]. The energy cutoff for the plane-wave expansion is 500 eV. The Brillouin zones are sampled by Monkhorst-Pack k-point meshes [24] for all compounds with meshes chosen to give a constant density of k points (30 Å^3). The geometry is relaxed until the total energy is less than 10^{-5} eV and the forces are below 0.01 eV/Å. The elastic constants ($\mathbf{C}$ or c_{ij}) are calculated from the strain-stress relationship [25]: $\sigma_\mathbf{s} = \mathbf{C} \cdot \epsilon$, where $\sigma_\mathbf{s}, \mathbf{C}$, and ϵ are the engineering stress vector, the stiffness matrix, and the strain vectors, respectively. According to the Voigt-Reuss-Hill (VRH) theory in a macroscopic system [26], the corresponding elastic properties, such as the bulk modulus B and shear modulus G, can be evaluated from the elastic constants. The VRH approach is a useful methodology to calculate the elastic modulus for cubic isotropic compounds, such as PbS, PbSe, and PbTe [19]. Even for the anisotropic compounds (such as MgO and CaF_2), the elastic moduli calculated with the VRH approach are still in good agreement with the experimental measurements [27]. Anderson [28] suggested that the VRH approximation is quite accurate after applying it to 200 crystals of different classes. Therefore, we will use the approach to calculate the elastic modulus.

The sound velocities of the longitude (v_L) and shear (v_S) waves and the corresponding averaged velocity v_a can be written as [28,29]

$$v_L = \sqrt{\frac{B + 4/3G}{\rho}}, \quad v_S = \sqrt{\frac{G}{\rho}},$$

$$v_a = \left[\frac{1}{3}\left(\frac{1}{v_L^3} + \frac{2}{v_S^3}\right)\right]^{-\frac{1}{3}}, \tag{2}$$

where ρ is the density of a compound. Additionally, the Debye temperature Θ_e can be expressed by using the sound velocity,

$$\Theta_e = \frac{h}{k_B}\left[\frac{3m}{4\pi}\right]^{\frac{1}{3}} v_a n^{-1/3}. \tag{3}$$

Here, h, k_B, and m are the Planck constant, Boltzmann constant, and number of atoms per volume. The Debye temperature calculated with the elastic properties characterizes the total vibrational spectrum. (Details are given in the Supplemental Material [30].) However, the lattice thermal conductivity calculation needs the Debye temperature only for acoustic branches. Thus, $n^{-1/3}$ in Eq. (3) is used to roughly separate the acoustic branches from the total vibration spectrum [16,31]. Since we use the primitive cell to carry out simulations, n is the number of atoms in the primitive cell.

III. ELASTIC PROPERTIES

For the 39 compounds with the RS, ZB, and WZ prototypes considered in our work, 26 compounds have experimentally measured elastic constants (see Table S1 in the Supplemental Material). These compounds include the well-studied thermoelectric materials: PbS, PbSe, PbTe, CdS, and CdSe. The corresponding experimental elastic moduli (the bulk modulus B and the shear modulus G) are estimated using the Voigt-Reuss-Hill approximation. The theoretically calculated elastic properties are in good agreement with the experimental measurements (see Table S1 in the Supplemental Material). This confirms that the theoretically calculated elastic properties are reasonable, and we will use the same computational methodology for the mechanical properties of compounds without experimental measurements.

In order to catch the general trend between the elastic property and the lattice thermal conductivity, the compounds in Table S1 in the Supplemental Material involve not only semiconductors but also some insulators (such as NaCl). We notice that for the same prototype, the mechanical properties (B and G) become smaller as the anion/cation radius becomes larger (the anion/cation goes down in the periodic table along a given column, e.g., $B_{\text{NaI}} < B_{\text{NaBr}} < B_{\text{NaCl}}$ and $B_{\text{PbTe}} < B_{\text{PbSe}} < B_{\text{PbS}}$, $B_{\text{RbCl}} < B_{\text{KCl}} < B_{\text{NaCl}}$ and $B_{\text{CdS}} < B_{\text{ZnS}}$). The larger the atomic radius is, the less electronegative (electropositive) the cation (anion) becomes. The soft mechanical properties with large radius are due to the strong Coulomb potential screening, resulting in the weak interatomic binding. The atoms in such a system (such as PbTe) can easily be deviated from their equilibrium positions with increasing temperature, which could lead to intriguing properties, such as the ferroelectric behavior in PbTe [32]. The weak atomic bond suggests strong anharmonicity and low lattice thermal conductivity, as discussed in Refs. [4,7,13], which is consistent with the experimentally measured thermal conductivity trend (see Table S2 in Supplemental Material): $\kappa_{\mathrm{NaI}} < \kappa_{\mathrm{NaBr}} < \kappa_{\mathrm{NaCl}}$, $\kappa_{\mathrm{PbTe}} < \kappa_{\mathrm{PbSe}} < \kappa_{\mathrm{PbS}}$, $\kappa_{\mathrm{RbCl}} < \kappa_{\mathrm{KCl}} \leqslant \kappa_{\mathrm{NaCl}}$, and $\kappa_{\mathrm{CdS}} < \kappa_{\mathrm{ZnS}}$.

IV. THERMAL PROPERTIES EVALUATED USING ELASTIC PROPERTIES

From the above calculations, we could qualitatively estimate the trend of lattice thermal conductivity using the elastic properties (B and G). However, to compare with the experimentally measured thermal conductivity, we need to quantitatively calculate the conductivity. To do so, we need the acoustic Debye temperature and Grüneisen parameter to calculate the lattice thermal conductivity [Eq. (1)]. As we mentioned before, the (quasi)phonon calculations are required to accurately estimate these thermal properties (Θ and γ). To release the computational burden, we alternatively use the elastic properties to efficiently obtain Θ and γ. For the acoustic Debye temperature, Θ_e [Eq. (3)] is used to approximate the accurate Debye temperature (such as the one determined using the phonon density state, Θ_ω).

A. Debye temperatures

The theoretically calculated Debye temperatures Θ_e of our 39 compounds with three prototype structures (RS, ZB, and WZ) are in good agreement with the phonon-determined Debye temperatures Θ_ω (Fig. 2 and Table S2 in the Supplemental Material). To further confirm the validation of Eq. (3), we choose one experimentally and theoretically well studied promising high-performance compound, SnSe [7], with the *Pnma* space group; its theoretically calculated bulk and shear moduli are 24.1 and 13.9 GPa, respectively. The corresponding averaged sound velocity [Eq. (2)] $v_a = 1718$ m/s, which is in good agreement with the phonon-calculated 1915 m/s (averaged velocities along all directions in the Brillouin zone from Ref. [7]). The Debye temperature of SnSe calculated using Eq. (3) is 84 K, which is in reasonable agreement with that determined using the phonon calculations (~60 K).

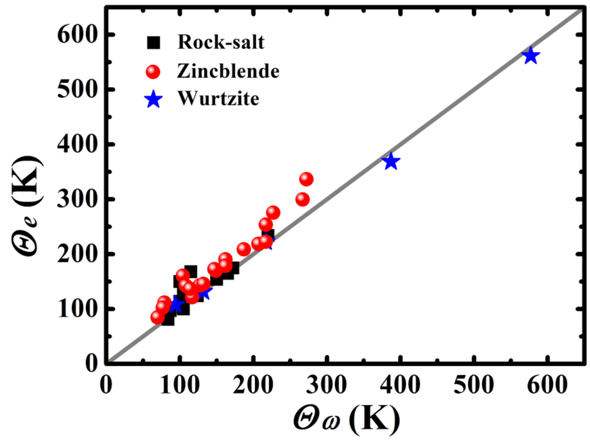

FIG. 2. Debye temperature determined by phonon density of states Θ_ω [15,17] versus the Debye temperatures determined by the elastic property Θ_e [Eq. (3)] for three prototype structures (RS: black squares; ZB: red dots; WZ: blue stars). The gray line is the ideal case of $\Theta_\omega = \Theta_e$. Data are given in the Supplemental Material (Table S2).

B. Grüneisen parameters: γ_ν

After treating the acoustic Debye temperature using the elastic properties, we turn to the acoustic Grüneisen parameter, or anharmonicity of a system. The sound velocity indicates the strength of interatomic interactions: the weaker the interaction is, the lower the sound velocity becomes, and the larger the Grüneisen parameter might be. Thus, for simplicity, the sound velocity presents a hint of anharmonicity strength (or the magnitude of the Grüneisen parameter). From previous works, the relationship between the Poisson ratio ν and the Grüneisen parameter can be expressed as [33–36]

$$\gamma_\nu = \frac{3}{2}\left(\frac{1+\nu}{2-3\nu}\right), \quad \nu = \frac{1-2(v_S/v_L)^2}{2-2(v_S/v_L)^2}. \tag{4}$$

Obviously, the sound-velocity-determined Grüneisen method is computationally more feasible than the quasiharmonic phonon calculations. Xiao *et al.* [19] used the formula to investigate the Grüneisen parameters γ_ν of four compounds (PbS, PbSe, PbTe, and SnSe) and found they are in good agreement with the quasiharmonic phonon-calculated results γ_ω. However, an obvious shortcoming of using Eq. (4) to determine the Grüneisen parameter is that it depends only on the ratio of v_S to v_L (v_S/v_L) and cannot describe the contributions from the absolute sound velocities (v_S or v_L). For two compounds (A and B), if the sound velocity of compound A is two times faster than that of compound B, compound B in principle has a large Grüneisen parameter. However, Eq. (4) results in the same unphysical Grüneisen parameter for the two compounds due to the same ratio of v_S to v_L or the Poisson ratio. Therefore, it is better to first check the suitability of the equation [Eq. (4)] for other compounds with different prototype structures (RS, ZB, and WZ; see Table S2 in the Supplemental Material). These compounds have Grüneisen parameters γ_ω determined by the lattice dynamic (or phonon; see Table S2 in the Supplemental Material). We notice that γ_ω of ZB and WZ are similar but clearly different from those of RS. This results from the different local geometries of the three prototypes (Fig. 1): There are six nearest neighbors (NN) in the RS structure but four NN in the ZB and WZ structures. Since the lattice dynamic property is dominated by the neighboring atoms or the number of bonds, the anharmonicity or the Grüneisen parameters of RS and ZB (or WZ) should be different. The explanation provides a useful guideline to roughly categorize the Grüneisen parameter according to the local geometry.

We then plot the Poisson-ratio-determined Grüneisen parameter γ_ν versus γ_ω for compounds with RS, ZB, and WZ structures [Fig. 3(a) and Table S2 in the Supplemental Material]. We can see that all the data in Fig. 3(a) split into two groups: One is for RS and the other is for ZB and WZ. For the compounds with the RS structure, γ_ν is

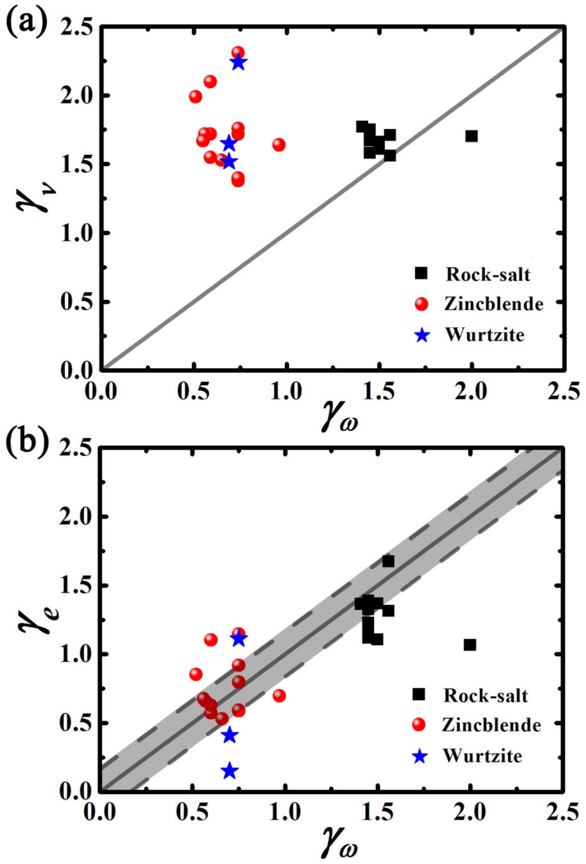

FIG. 3. Grüneisen parameter determined (a) using the Poisson ration γ_ν [Eq. (4)] and (b) using the B/G change with volume γ_e [Eq. (5)] for three prototype structures (RS, ZB, and WZ) versus the Grüneisen parameter determined using the lattice dynamic calculations γ_ω. The gray region is the γ_e error bar (0.23). Data are given in the Supplemental Material (Table S2).

in excellent agreement with γ_ω. This explains the reasonable Grüneisen parameters γ_ν of PbS, PbSe, and PbTe presented in Ref. [19] since these compounds belong to the RS structure. However, for the other prototype structures (ZB and WZ), the Poisson-ratio-determined Grüneisen parameters γ_ν are far above γ_ω, indicating Eq. (4) is not a general method for all compounds. This agrees with the conclusion of Belomestnykh [35] that Eq. (4) is oversimplified to use to estimate the Grüneisen parameters for all compounds. It is necessary to establish a distinct methodology to efficiently and accurately estimate the Grüneisen parameter.

C. Grüneisen parameters: γ_e

We have to return to the original Grüneisen parameter definition. The Grüneisen parameter γ_i characterizes the relationship between phonon frequency ω_i and volume V change and can be defined as [37]

$$\gamma_i = -\frac{V}{\omega_i}\frac{\partial \omega_i}{\partial V}. \tag{5}$$

The phonon frequency ω_i is proportional to the sound velocity v_i in the long-wave limit (or at the center of Brillouin zone): $\omega_i = v_i q$, where q is the wave vector. Substituting ω_i and the longitude and shear sound velocities [Eq. (2)] into Eq. (5), we can derive the equations for the longitude (γ_L^e), shear (γ_S^e), and averaged (γ_e) acoustic Grüneisen parameters:

$$\begin{aligned}
\gamma_L^e &= -\frac{\partial ln\omega_L}{\partial lnV} = -\frac{\partial v_L|q|}{\partial lnV} = -\frac{1}{2}\frac{V}{B+\frac{4G}{3}}\frac{\partial\left(B+\frac{4G}{3}\right)}{\partial V} - \frac{1}{6},\\
\gamma_S^e &= -\frac{\partial ln\omega_S}{\partial lnV} = -\frac{\partial v_S|q|}{\partial lnV} = -\frac{1}{2}\frac{V}{G}\frac{\partial G}{\partial V} - \frac{1}{6},\\
\gamma_e &= \sqrt{\left[\left(\gamma_L^e\right)^2 + 2\left(\gamma_S^e\right)^2\right]/3},
\end{aligned} \tag{6}$$

where $\frac{\partial(|q|\rho^{1/2})}{\partial lnV} = \frac{1}{6}$. Thus, from our derived acoustic Grüneisen formula, γ_e characterizes the relationship between the elastic properties (B and G) and volume V change. To evaluate γ_e, we compress the volume of a system by 4% and calculate the change in the elastic properties with volume. [In Table S3 in the Supplemental Material, we choose different volume changes (3%–6%) and find that the corresponding Grüneisen parameter is just slightly different.]

From the theoretically calculated γ_e of all three prototype compounds (RS, ZB, and WZ; Table S2 in Supplement Material), the Grüneisen parameter γ_e determined from the elastic properties (B and G) is in reasonable agreement with the phonon-determined result [Fig. 3(b)]. This is unlike the Poisson-ratio-determined Grüneisen parameter [γ_ν in Eq. (4)], which is suitable only for the RS structure [Fig. 3(a)]. For two compounds (PbS and GaN), their γ_e are different from γ_ω, which might be due to an optical-mode-influenced B/G change with volume or a strong electron correlation effect (such as in GaN). The evaluated error bar of γ_e is $\Delta\gamma_e = \frac{1}{N}\sum_{i=1}^{N}|\gamma_\omega^i - \gamma_e^i| = 0.23$ [the gray region in Fig. 3(b)]. The agreement between γ_e and γ_ω is because B and G correctly average the important acoustic branches around the center of the Brillouin zone, and Eq. (6) captures the important frequency change with volume. All compounds of the three prototypes are based on high-symmetry lattices (the cubic isotropic RS and ZB and the noncubic anisotropic WZ). Without loss of generality, we further apply our B/G-determined Grüneisen parameter method [Eq. (6)] to the noncubic anisotropic layered SnSe compound, and the calculated $\gamma_e = 3.32$ is consistent with the quasiharmonic phonon-calculated value of 3.26. Particularly, the B/G calculation is computationally cheaper than the quasiharmonic approximation. Therefore, Eq. (6) can be used as an efficient anharmonicity or Grüneisen parameter evaluation tool.

D. Lattice thermal conductivity

Inserting the acoustic Debye temperature Θ_e and Grüneisen parameter γ_e determined using the elastic properties (B and G) into the lattice thermal conductivity formula performed using the Slack model [Eq. (1)], we can calculate the lattice thermal conductivity κ_e of all compounds with the three prototype structures at $T = 300$ K (see Table S2 in the Supplemental Material). From the experimental measurements [17], these compounds exhibit a wide range of lattice thermal conductivity behavior, 0.4–350 W/mK at 300 K. The experimentally measured thermal conductivity is the total conductivity, including the lattice thermal conductivity and electronic thermal conductivity. For the compounds we considered (semiconductors

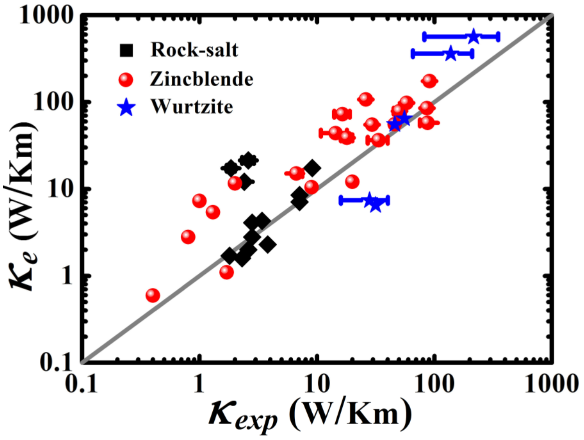

FIG. 4. Theoretically calculated lattice thermal conductivity κ_e [Eq. (1)] using the elastic properties (B and G) versus the experimental measurement κ_{exp}. The error bars on some points are considering the experimental measurement uncertainty. Data are given in the Supplemental Material (Table S2). The logarithmic scale is used to clearly represent the plot.

and insulators all having low carrier densities), the electronic contribution is so small that it can be ignored. Therefore, it is reasonable to compare the theoretically calculated lattice thermal conductivity with the experimentally measured total thermal conductivity. The theoretically calculated κ_e are in reasonable agreement with the experimental measurements (κ_{exp} at $T = 300$ K) in the whole thermal conductivity range (see Fig. 4), and this trend is consistent with the one solving the full solution of the linearized phonon Boltzmann equation [20]. For several compounds (such as ZnS), the large difference between κ_e and κ_{exp} could possibly result from (a) the optical branch contributions in the thermal conductivity, which are not considered in the Slack model [Eq. (1)]; (b) the defect contributions, which occur in the experimentally measured samples but not in the theoretically used perfectly ordered crystalline; or (c) the possible experimentally measured uncertainty. Additionally, for the noncubic anisotropic SnSe compound, the elastic-property-determined lattice thermal conductivity κ_e is 0.2 W/mK, which is in reasonable agreement with the experimental measurement (∼0.5 W/mK) and is in good agreement with that (∼0.3 W/mK) calculated using the first-principles Debye-Callaway approach [7].

IV. CONCLUSIONS

We have established a methodology that can be used to calculate the lattice thermal conductivity using the computationally feasible elastic properties (the bulk and shear moduli) and applied the method to 39 binary compounds with three different prototypes (including the cubic isotropic rocksalt and zinc blende and the noncubic anisotropic wurtzite). The theoretically calculated Debye temperature, Grüneisen parameter, and lattice thermal conductivity are in good agreement with those determined using the (quasi)harmonic phonon calculations or the experimental measurements. However, the elastic properties (such as the bulk and shear moduli) average the elastic constants only along certain directions ($C_{ij}, i, j = x, y,$ and z). C_{ij} cannot consider all high-symmetry directions in the Brillouin zone as phonon calculations do. Thus, some important acoustic contributions might be missed in the approach. This might specifically lead to a large error for strongly anisotropic systems with low-symmetry lattices (such as the triclinic lattice) plus unusual atomic arrangements (such as forming a large void space). Further work is necessary to understand and overcome the limitation. Nevertheless, the soft mechanical properties induce the low Debye temperature and strong anharmonicity or large Grüneisen parameter due to the easy deformation of crystal structures under stress. Our method provides an efficient, feasible (computationally cheaper) way to estimate the anharmonicity and lattice thermal conductivity and has benefits for seeking not only low-thermal-conductivity compounds for high-performance thermoelectric materials but also high-thermal-conductivity compounds for thermally conductive materials.

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[30] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.95.155206 for the Debye temperature calculation using elastic properties, and the theoretically calculated and experimentally measured thermal properties (the Debye temperature, Grüneisen parameter and lattice thermal conductivity).

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