High-throughput computational screening of thermal conductivity, Debye temperature, and Grüneisen parameter using a quasiharmonic Debye model

Cormac Toher, Jose J. Plata, Ohad Levy, Maarten de Jong, Mark Asta, Marco Buongiorno Nardelli, and Stefano Curtarolo

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I. INTRODUCTION

Calculating the thermal properties of materials is important for predicting the thermodynamic stability of structural phases and assessing their importance for a variety of applications. The lattice thermal conductivity, $\kappa_l$, is a crucial design parameter in a wide range of important technologies, such as the development of new thermoelectric materials, heat sink materials for thermal management in electronic devices, and rewritable density scanning-probe phase-change memories. High thermal conductivity materials, which typically have a zinclende or diamond-like structure, are essential in microelectronic and nanoelectronic devices to achieve efficient heat removal, and have been intensively studied for the past few decades. Low thermal conductivity materials constitute the basis of a new generation of thermoelectric materials and thermal barrier coatings.

The determination of the thermal conductivity of materials is computationally demanding since it requires calculation of multiple-phonon scattering processes, that are the origin of the lattice resistance to heat transport. The methods most commonly used currently to calculate the thermal conductivity are based on solving the Boltzmann Transport Equation (BTE). This solution involves the calculation of the phonon frequencies, group velocities, and the harmonic and anharmonic interatomic force constants (IFCs). In particular, the third-order anharmonic IFCs are required in order to incorporate the effects of three phonon scattering processes. The standard method to calculate these anharmonic IFCs is based on density functional theory (DFT). Deinzer et al. used Density Functional Perturbation Theory (DFPT) to obtain third-order IFCs to study the phonon linewidths. In the last decade, this method has been successfully used to solve the BTE and predict the thermal conductivity of different materials. Such evaluation of the higher-order IFCs requires electronic structure calculations for multiple large supercells, each of which has a different set of atomic displacements. These first principles solutions of the BTE are therefore computationally extremely expensive.

A variety of simple methods have been devised to evaluate the thermal properties of materials at reduced computational cost. Early approximate implementations to compute the lattice thermal conductivity were based on semi-empirical models to solve the BTE, in which some parameters are obtained from fitting to experimental data. This reduces the predictive power of the calculations.

An alternative approach to calculating thermal conductivity is based on the Green-Kubo formula, which employs molecular dynamics simulations to calculate thermal currents over long time periods after thermal equilibrium is reached. This technique takes into account high order scattering processes, but the usage of semi-empirical potentials leads to errors on the order of 50%.

The methods described above are unsuitable for rapid generation and screening of large databases of materials properties in order to identify trends and simple descriptors for thermal properties. To accomplish this, we chose to implement a much cheaper approach, the “GIBBS” quasi-harmonic Debye model. This approach does not require large supercell calculations since it relies merely on first-principles calculations of the energy as a function of unit cell volume. It is thus much more tractable computationally and is eminently suited to investigating the thermal properties of entire classes of materials.
Helmholtz vibrational energy is melting point. In the quasi-harmonic approximation, the most materials at temperatures significantly below their free energy is small, which is a good approximation for and intrinsic point defect contributions to the Helmholtz framework) and the Materials Project\textsuperscript{27–29} frameworks for high-throughput computational materials science, and use it to construct a database of computed compound thermal conductivities and Debye temperatures.

\section{II. The Automatic Gibbs Library (AGL)}

The AGL software library implements the “GIBBS” method\textsuperscript{23} in the AFLLOW\textsuperscript{24–26} framework (C++ based framework) and the Materials Project\textsuperscript{27–29} (Python implementation). The library includes automatic error handling and correction to facilitate high-throughput computation of materials thermal properties. The principal ingredients of the calculation are described in the following sections.

\subsection{A. The GIBBS quasi-harmonic Debye model}

In thermodynamics, the equilibrium state of a system at a constant temperature and pressure minimizes its Gibbs free energy

\[ G(\vec{x}; p, T) = E(\vec{x}) + pV(\vec{x}) + A_{\text{vib}}(\vec{x}; T), \]  

where \( \vec{x} \) is a configuration vector containing all the information about the system’s geometry, \( E(\vec{x}) \) is the total energy of the crystal (obtained, for example, from an electronic structure calculation), \( A_{\text{vib}} \) is the vibrational Helmholtz free energy, and \( p \) and \( V(\vec{x}) \) are the pressure and volume. It is assumed here that the electronic and intrinsic point defect contributions to the Helmholtz free energy is small, which is a good approximation for most materials at temperatures significantly below their melting point. In the quasi-harmonic approximation, the Helmholtz vibrational energy is

\[ A_{\text{vib}}(\vec{x}; T) = \int_0^\infty \left[ \frac{\hbar \omega}{2} + k_B T \log \left( 1 - e^{-\hbar \omega/k_B T} \right) \right] g(\vec{x}; \omega) d\omega, \]  

\[ \text{(2)} \]

where \( g(\vec{x}; \omega) \) is the phonon density of states. As mentioned before, calculation of the full phonon density of states is computationally demanding, requiring electronic structure calculations for multiple supercell configurations. Instead, the “GIBBS” method uses a quasi-harmonic Debye model, where the Helmholtz free energy is expressed in terms of the Debye temperature \( \theta_D \)

\[ A_{\text{vib}}(\theta_D; T) = n k_B T \left[ \frac{9}{8} \frac{\theta_D}{T} + 3 \log \left( 1 - e^{-\theta_D/T} \right) \right] - D \left( \frac{\theta_D}{T} \right), \]  

\[ \text{(3)} \]

where \( n \) is the number of atoms in the unit cell and \( D(\theta_D/T) \) is the Debye integral

\[ D \left( \frac{\theta_D}{T} \right) = \frac{T}{\theta_D} \int_0^{\theta_D/T} \frac{x^3}{e^x - 1} dx. \]  

\[ \text{(4)} \]

In isotropic solids, changes in the geometry can be treated as isotropic changes in the volume, such that the magnitude of the configurational vector \( \vec{x} \) is equal to the cube root of the volume, i.e. \( x = V^{1/3} \). The value of \( \theta_D \) can thus be calculated as\textsuperscript{23,30,31}

\[ \theta_D = \frac{\hbar}{k_B} [6\pi^2 V^{1/3} n]^{1/3} f(\sigma) \sqrt{B_S / M}. \]  

\[ \text{(5)} \]

Here, \( M \) is the mass of the unit cell, \( B_S \) is the adiabatic bulk modulus, and \( f(\sigma) \) is given by

\[ f(\sigma) = \left\{ \begin{array}{ll} 3 & \left( \frac{2}{3} \frac{1+\sigma}{3} \right)^{3/2} + \left( \frac{1+\sigma}{3} \frac{1+\sigma}{3} \right)^{3/2} \end{array} \right\}^{-1/3}, \]  

\[ \text{(6)} \]

in the assumption that the Poisson ratio \( \sigma \) is constant. The value of the Poisson ratio can be set as an input to AGL separately from the DFT calculations, e.g., to the experimentally measured value. For the calculations described in this paper this value is set at 0.25, which is the theoretical value for a Cauchy solid\textsuperscript{23,31}. The Poisson ratio \( \sigma \) for crystalline materials is typically in the range of 0.2 to 0.3. Since the function \( f(\sigma) \) behaves approximately linearly with values running from 0.9 to 0.7 when \( \sigma \) is in the range from 0.2 to 0.3, this approximation has only a small effect on the results. We have checked this by performing the AGL calculations using the literature values of the Poisson ratio where they are available. The correlation between calculated and experimental values of the thermal conductivity is typically within a few percent of that obtained with the constant value of 0.25.

The adiabatic bulk modulus, \( B_S \), can be approximated by the zero temperature limit of the isothermal compressibility (neglecting zero-point contributions), which we will refer to as \( B_{\text{r_static}} \):

\[ B_S \approx B_{\text{static}}(\vec{x}) \approx B_{\text{static}}(\vec{x}_{opt}(V)) = V \left( \frac{\partial^2 E(\vec{x}_{opt}(V))}{\partial V^2} \right) = V \left( \frac{\partial^2 E(V)}{\partial V^2} \right), \]  

\[ \text{(7)} \]

where \( \vec{x}_{opt} \) is the configuration vector of the unit cell geometry. The Gibbs free energy of the system can be expressed as a function of the unit cell volume

\[ G(V; p, T) = E(V) + pV + A_{\text{vib}}(\theta_D(V); T), \]  

\[ \text{(8)} \]

where \( \theta_D \) as a function of volume is evaluated from Equations (5) and (7), and \( E(V) \) is obtained from a set of DFT calculations for unit cells with different volumes. Minimizing the Gibbs free energy with respect to volume, the equilibrium configuration at \((p, T)\) is determined, and additional properties, including the equilibrium \( \theta_D \), bulk modulus, heat capacity, thermal coefficient of expansion, etc. can be evaluated.
B. Thermal calculation procedure

In order to calculate the thermal properties for a particular material with a particular structure, first a set of DFT (e.g. VASP\textsuperscript{32}) calculations which only differ by isotropic variations in the unit cell volume are set up and run using the AFLLOW or Materials Project framework. The resulting $E(V)$ is fitted by a polynomial, to calculate the adiabatic bulk modulus, $B_s$, as a function of volume from Equation (7). The $B_s$ values are then used to calculate the Debye temperature $\theta_D$ for each unit cell volume from Equation (5). Next, the vibrational Helmholtz free energy $A_{vib}(\theta_D(V); T)$ as a function of volume, is calculated using Equation (3) for a given value of the temperature, $T$. The zero-pressure GIBBS free energy as a function of volume is then obtained by

$$G(x; 0, T) = E(V) + A_{vib}(\theta_D(V), T). \quad (9)$$

This Gibbs free energy is fitted by a polynomial which is minimized with respect to volume to find the equilibrium volume for any given value of $T$, at zero pressure. The polynomial is an expansion in $x = \sqrt{V}$. Therefore, finite pressure can be added simply to the coefficient of the $x^3$ term. The volume which minimizes this modified polynomial for $G(p, V, T)$ is the equilibrium volume that gives the Gibbs free energy for each requested $(p, T)$. This equilibrium volume is used to calculate the bulk modulus and Debye temperature of the material as a function of $p$ and $T$, from Equations (7) and (5), respectively.

C. DFT calculation details

The DFT calculations to obtain $E(V)$ were performed using the VASP software\textsuperscript{32} with projector-augmented-wave pseudopotentials\textsuperscript{33} and the PBE parameterization of the generalized gradient approximation to the exchange-correlation functional\textsuperscript{34}. The energies were calculated at zero temperature and pressure, with spin polarization and without zero-point motion or lattice vibrations. The initial crystal structures were fully relaxed (cell volume and shape and the basis atom coordinates inside the cell). An additional 100 different volume cells were calculated for each structure by increasing or decreasing the relaxed lattice parameter in fractional increments of 0.005. Numerical convergence to about 1 meV/atom was ensured by a high-energy cut-off (30% higher than the maximum cutoff of each of the potentials) and a 8000 k-point Monkhorst-Pack\textsuperscript{35} or Γ-centred (in the case of hexagonal unit cells) mesh.

D. The Grüneisen Parameter

The Grüneisen parameter describes how the thermal properties of a material vary with the unit cell size. It is defined by the phonon frequencies dependence on the unit cell volume

$$\gamma_i = \frac{V}{\omega_i} \frac{\partial \omega_i}{\partial V}. \quad (10)$$

Debye’s theory assumes that all mode frequencies vary with the volume in the same ratio as the cut-off frequency (Debye frequency), so the Grüneisen parameter can be expressed in terms of $\theta_D$

$$\gamma = -\frac{\partial \log(\theta_D(V))}{\partial \log V}, \quad (11)$$

and calculated using the Mie-Grüneisen equation\textsuperscript{31}

$$p - p_{T=0} = \gamma \frac{U_{vib}}{V}, \quad (12)$$

where $U_{vib}$ is the vibrational internal energy

$$U_{vib} = n k_B T \left[ \frac{9 \theta_D}{8 T} + 3 D \left( \frac{\theta_D}{T} \right) \right]. \quad (13)$$

The expression in Eq. (10) can also be related to the macroscopic definition via a weighted average with the heat capacities for each branch of the phonon spectrum

$$\gamma = \sum_i \gamma_i C_{V,i} \sum_i C_{V,i}. \quad (14)$$

that leads to the thermodynamic relations

$$\gamma = V \left( \frac{\partial P}{\partial E} \right)_V = \frac{\alpha B_s}{C_{pp}} = \frac{\alpha B_T}{C_V \rho}, \quad (15)$$

where $\rho$ is the density of the material.

An alternative expression for the Grüneisen parameter was derived by Slater under the assumption of a constant Poisson ratio\textsuperscript{36}

$$\gamma = -\frac{2}{3} - \frac{V}{2} \frac{d^2 p/dV}{dp/dV}. \quad (16)$$

Equations (11), (12), and (16) have all been implemented within the AGL. Unless otherwise specified, the values of the Grüneisen parameter listed in the results and used to calculate the thermal conductivity are obtained using Equation (12), as this is generally considered more accurate than Equation (11)\textsuperscript{23}.

E. Thermal conductivity

In the AGL, the thermal conductivity is calculated by the method proposed by Slack\textsuperscript{37,38} using the Debye temperature and the Grüneisen parameter

$$\kappa_i(\theta_a) = \frac{0.849 \times 3 \sqrt{3}}{20 \pi^3 (1 - 0.514 \gamma^{-1} + 0.228 \gamma^{-2})} \times \left( \frac{k_B \theta_a}{\hbar} \right)^2 \frac{k_B m V^{1/2}}{\hbar \gamma^2}. \quad (17)$$
where \( V \) is the volume of the unit cell and \( m \) is the average atomic mass. It should be noted that the Debye temperature in this formula, \( \theta_a \), is slightly different than the traditional Debye temperature, \( \theta_D \), calculated in Equation (5). Instead, \( \theta_a \) is obtained by only considering the acoustic modes, based on the assumption that the optical phonon modes in crystals do not contribute to heat transport\(^{37} \). This \( \theta_a \) is referred to as the “acoustic” Debye temperature\(^{37,38} \). It can be derived directly from the phonon DOS by integrating only over the acoustic modes\(^{37,39} \). Alternatively, it can be calculated from the traditional Debye temperature \( \theta_D \)\(^{37,38} \)

\[
\theta_a = \theta_D n^{-\frac{3}{4}}. \tag{18}
\]

To demonstrate the distinction between these two quantities, we include the values of both \( \theta_D \) and \( \theta_a \), as calculated using AGL, in the tables of results in the following sections.

The thermal conductivity at temperatures other than \( \theta_a \) is estimated by\(^{37,38,40} \):

\[
\kappa_i(T) = \kappa_i(\theta_a) \frac{\theta_a}{T}. \tag{19}
\]

In principle, the Grüneisen parameter in Equation (17) should also be derived only from the acoustic phonon modes\(^{37} \). However, unlike the case of \( \theta_D \) and \( \theta_a \), there is no simple way to extract it from the traditional Grüneisen parameter. Instead, it must be calculated from Equation (10) for each phonon branch separately and summed over the acoustic branches. This requires calculating the full phonon spectrum for different volumes, and is therefore too computationally demanding to be used for high-throughput screening. The dependence of the expression (17) on \( \gamma \) is weak\(^{38} \), thus the evaluation of \( \kappa_i \) using the traditional Grüneisen parameter introduces just a small systematic error which is insignificant for screening purposes.

### F. Pearson and Spearman Correlations

Pearson and Spearman correlations have been implemented separately from AGL, in order to analyze the results for entire sets of materials. The Pearson correlation coefficient \( r \) is a measure of the linear correlation between two variables, \( X \) and \( Y \). It is calculated by

\[
r = \frac{\sum_{i=1}^{n} (X_i - \overline{X})(Y_i - \overline{Y})}{\sqrt{\sum_{i=1}^{n} (X_i - \overline{X})^2 \sum_{i=1}^{n} (Y_i - \overline{Y})^2}}, \tag{20}
\]

where \( \overline{X} \) and \( \overline{Y} \) are the mean values of \( X \) and \( Y \).

The Spearman rank correlation coefficient \( \rho \) is a measure of the monotonicity of the relation between two variables. The raw values of the two variables \( X_i \) and \( Y_i \) are sorted in ascending order, and are assigned rank values \( x_i \) and \( y_i \), which are equal to their position in the sorted list. If there is more than one variable with the same value, the average of the position values are assigned to each. The correlation coefficient is then given by

\[
\rho = \frac{\sum_{i=1}^{n} (x_i - \overline{x})(y_i - \overline{y})}{\sqrt{\sum_{i=1}^{n} (x_i - \overline{x})^2} \sqrt{\sum_{i=1}^{n} (y_i - \overline{y})^2}}. \tag{21}
\]

It is useful for determining how well the ranking order of the values of one variable predict the ranking order of the values of the other variable.

### III. RESULTS

We used the AGL to calculate the the Debye temperature, Grüneisen parameter and thermal conductivity for a set of 75 materials with the diamond, zincblende, rocksalt and wurtzite structures, and 107 half-Heusler compounds. The results have been compared to first-principles calculations (and experimental values where available) of the half-Heusler compounds and to experimental values for the other structures.

#### A. Zincblende and Diamond structure materials

Experimental values of thermal properties for materials with the Zincblende (spacegroup: \( F43m \) (#216); Pearson symbol: \( cF8 \)) and Diamond (spacegroup: \( Fd3m \) (#227); Pearson symbol: \( cF8 \)) structures were published in Table II of Ref. 37 and Table 2.2 of Ref. 38. They are shown with the calculated thermal conductivity at 300K, the Debye temperature and Grüneisen parameter for these materials in Table I and in Figure 1. As shown in the table, for a few of these materials there are discrepancies in experimental values quoted in the different sources. For each entry we used the value from the most recent source for plotting the following figures and calculating the correlations reported here.

Comparison of the calculated and experimental results of Table I shows that the absolute agreement between them is quite poor, with discrepancies of tens, or even hundreds, of percent quite common. Considerable disagreements also exist between different experimental reports of these properties, in almost all cases where they exist. Unfortunately, the scarcity of experimental data from different sources on the thermal properties of these materials prevents reaching definite conclusions regarding the true values of these properties. The available data can thus only be considered as a rough indication of their order of magnitude.

Nevertheless, the Pearson correlation between the AGL calculated thermal conductivity values and the experimental values is high, 0.878. The Spearman correlation, 0.905, is even higher. The Spearman correlation between the experimental values of the thermal conductivity and \( \theta_a \) as calculated with AGL is 0.925. There is also a strong correlation between the experimental values of \( \theta_a \) and those calculated with AGL, with a Pearson correlation of 0.995.
and a Spearman correlation of 0.984. The correlation for the Grüneisen parameter is much worse, with Pearson and Spearman correlations of 0.137 and −0.187, respectively.

Table 2.2 of Ref. 38 includes values of the thermal conductivity at 300K, calculated using the experimental values of $\theta_a$ and $\gamma$. The Pearson correlation between these calculated thermal conductivity values and the experimental values is 0.932, and the corresponding Spearman correlation is 0.941. Both values are just slightly higher than the correlations we calculated using the AGL evaluations of $\theta_a$ and $\gamma$. Thus, the unsatisfactory quantitative reproduction of these quantities by the Debye quasi-harmonic model has little impact on its effectiveness as a screening tool for high or low thermal conductivity materials. The model can be used when these experimental values are unavailable.

These results indicate that despite the quantitative disagreement between the calculated and experimental results for the thermal conductivity and $\theta_a$, the AGL calculations are good indicators for the relative values of these quantities and for ranking materials in order of increasing conductivity. For the Diamond and Zincblende structure materials, the calculated $\theta_a$ turns out to be a slightly better indicator of the ordinal order of the thermal conductivity than the calculated conductivity.

### B. Rocksalt structure materials

Experimental values of the thermal properties of materials with the Rocksalt structure (spacegroup: $Fm\bar{3}m$ (#225); Pearson symbol: $cF8$) were published in

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**TABLE I.** Lattice thermal conductivity at 300K, Debye temperature and Grüneisen parameter of Zincblende and Diamond structure semiconductors. The values listed for $\Theta^{\exp}$ are $\theta_a$, except 141K for HgTe which is $\theta_D^{\exp}$. Units: $\theta$ in (K), $\kappa$ in (W/(m·K)).

| Comp. | $\Theta^{\exp}$ | $\theta_A^{AGL}$ | $\theta_C^{AGL}$ | $\gamma^{\exp}$ | $\gamma_A^{AGL}$ | $\kappa^{\exp}$ | $\kappa_A^{AGL}$ |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|----------------|-----------------|
| C     | 1450$^{37,38}$ | 1219            | 1536            | 0.75$^{37}$    | 1.74            | 3000$^{37}$   | 1691           |
| Si    | 740$^{37}      | 737             | 928             | 0.76$^{37}$    | 1.84            | 360$^{41}$    | 67.19          |
| Ge    | 235$^{37,38}$  | 235             | 296             | 1.06$^{38}$    | 2.09            | 166$^{38}$    | 20.58          |
| BN    | 1200$^{38}$    | 1118            | 1409            | 0.73$^{38}$    | 1.73            | 760$^{38}$    | 138.38         |
| BP    | 670$^{37,38}$  | 644             | 811             | 0.75$^{38}$    | 1.78            | 350$^{38}$    | 52.56          |
| AlP   | 381$^{38}$     | 430             | 542             | 0.75$^{38}$    | 1.96            | 90$^{12,43}$  | 21.16          |
| AlAs  | 270$^{37,38}$  | 300             | 378             | 0.66$^{37,38}$ | 2.04            | 98$^{38}$     | 12.03          |
| AlSb  | 210$^{37,38}$  | 223             | 281             | 0.63$^{37,38}$ | 2.12            | 56$^{38}$     | 7.22           |
| GaP   | 275$^{37,38}$  | 314             | 396             | 0.75$^{38}$    | 2.15            | 100$^{38}$    | 11.76          |
| GaAs  | 220$^{37,38}$  | 240             | 302             | 0.75$^{37,38}$ | 2.23            | 45$^{38}$     | 7.2            |
| GaSb  | 165$^{37,38}$  | 186             | 234             | 0.75$^{37,38}$ | 2.27            | 46$^{38}$     | 4.62           |
| InP   | 220$^{37,38}$  | 241             | 304             | 0.65$^{37,38}$ | 2.22            | 93$^{38}$     | 7.78           |
| InAs  | 165$^{37,38}$  | 195             | 246             | 0.57$^{37,38}$ | 2.26            | 30$^{38}$     | 5.36           |
| InSb  | 135$^{37,38}$  | 158             | 199             | 0.56$^{37,38}$ | 2.3             | 20$^{38}$     | 3.64           |
| ZnS   | 290$^{37,38}$  | 301             | 379             | 0.75$^{37,38}$ | 2.01            | 27$^{38}$     | 11.33          |
| ZnSe  | 190$^{37,38}$  | 230             | 290             | 0.75$^{37,38}$ | 2.07            | 19$^{38}$     | 7.48           |
| ZnTe  | 155$^{37,38}$  | 181             | 228             | 0.97$^{37,38}$ | 2.14            | 18$^{38}$     | 4.87           |
| CdSe  | 103$^{38}$     | 186             | 234             | 0.63$^{38}$    | 2.19            | 4.47          | 4.99           |
| CdTe  | 120$^{37,38}$  | 152             | 191             | 0.52$^{37,38}$ | 2.23            | 7.5$^{38}$    | 3.49           |
| HgSe  | 110$^{37}$     | 151             | 190             | 0.17$^{37}$    | 2.4             | 344           | 3.22           |
| HgTe  | 141$^{37}$     | 129             | 162             | 1.97$^{37}$    | 2.46            | 2.5$^{37}$    | 2.36           |

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**FIG. 1.** (a) Lattice thermal conductivity at 300K, (b) acoustic Debye temperature and (c) Grüneisen parameter of Zincblende and Diamond structure semiconductors.
Table III of Ref. 37 and Table 2.1 of Ref. 38. They are compared to the values calculated by the AGL in Table II and Figure 2. As was the case for the zincblende structure materials, we have included the AGL results for both $\theta_D$ and $\theta_a$ in the table. The experimental values listed in the table are all for $\theta_D$ except for SnTe where $\theta_D$ is quoted in Ref. 7. The AGL $\theta_a$ values were used for plotting and correlation calculations, with the exception of that for SnTe where $\theta_D$ was used for plotting Figure 2b and for calculating the correlation between the Debye temperatures.

The Pearson correlation between the calculated and experimental values for the thermal conductivity is 0.91. The Spearman correlation is 0.445. The Spearman correlation between the experimental values of the thermal conductivity and the calculated values of $\theta_a$ is 0.645. The Pearson correlation between the calculated and experimental values for the Debye temperature is 0.982 and the corresponding Spearman correlation is 0.948. The correlation for the Gruneisen parameter is much worse, with Pearson and Spearman correlations of 0.118 and −0.064, respectively.

Table 2.1 of Ref. 38 includes values of the thermal conductivity at 300K, calculated using the experimental values of $\theta_a$ and $\gamma$. The Pearson correlation between these calculated thermal conductivities and their experimental values is 0.986, and the corresponding Spearman correlation is 0.761. Comparing these values with the correlations obtained using the AGL calculated quantities, we find that the latter are more significantly degraded than for the Diamond and Zincblende structures. This is despite the similar correlations obtained for $\theta_a$ and $\gamma$ in these two cases. Nevertheless, the Pearson correlation between the calculated and experimental conductivities is high in both calculations, indicating that the AGL approach may
be used as a screening tool for high conductivity compounds in cases where gaps exist in the experimental data for these materials.

C. Wurzite structure materials

Experimental results for Wurzite structure materials (spacegroup: P63mc (#186); Pearson symbol: hP4) appear in Table 2.3 of Ref. 38. Their comparison with our calculation results is shown in Table III and Figure 3. As was the case for the zincblende and wurzite structure materials, we have included the AGL results for both \( \theta_D \) and \( \theta_a \) in the table, while the AGL \( \theta_a \) was used for plotting Figure 3 and calculating the correlations. The experimental values listed in the table are all for \( \theta_a \), with the exceptions of the values of 190K for InSe\(^7\) and 660K for InN\(^{41,46}\), which are for \( \theta_D \). The AGL \( \theta_a \) values were used for plotting and correlation calculations, with the exception of that those for InSe and InN where \( \theta_D \) was used for plotting Figure 3b and for calculating the correlation between the Debye temperatures.

The Pearson correlation between the AGL thermal conductivity values and the experimental values is 0.943. The corresponding Spearman correlation is 0.976. The Spearman correlation between the experimental values of the thermal conductivity and the calculated values of \( \theta_a \) is 0.905. The Pearson correlation between the experimental and calculated values of the Debye temperature is 0.8, and the corresponding Spearman correlation is 0.833. The correlations for the Grüneisen parameter are both poor, with Pearson and Spearman values of \(-0.039\) and \(0.160\), respectively.

Table 2.3 of Ref. 38 includes values of the thermal conductivity at 300K, calculated using the experimental values of the Debye temperature and Grüneisen parameter. The Pearson correlation between these calculated thermal conductivity values and the experimental values is 0.996, and the corresponding Spearman correlation is 1.0. These values are again higher than the correlations obtained using the AGL calculated quantities, however, all of these correlations are very high so either of the calculation methods could serve as a reliable screening tool of the thermal conductivity. It should be noted that the high correlations calculated with the experimental \( \theta_a \) and

| Comp. | \( \theta_a^{\text{exp}} \) | \( \theta_a^{\text{AGL}} \) | \( \gamma^{\text{exp}} \) | \( \gamma^{\text{AGL}} \) | \( \kappa^{\text{exp}} \) | \( \kappa^{\text{AGL}} \) |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| SiC   | 740\(^{38}\)   | 750             | 1191            | 0.738           | 1.86            | 490             |
|       |                 |                 |                 |                 | 52.63           |                 |
| AlN   | 620\(^{38}\)   | 485             | 770             | 0.738           | 1.85            | 350             |
|       |                 |                 |                 |                 | 32.58           |                 |
| GaN   | 390\(^{38}\)   | 291             | 462             | 0.738           | 2.07            | 210             |
|       |                 |                 |                 |                 | 14.55           |                 |
| ZnO   | 303\(^{38}\)   | 519             | 824             | 0.738           | 1.97            | 60              |
|       |                 |                 |                 |                 | 20.98           |                 |
| BeO   | 800\(^{38}\)   | 784             | 1244            | 1.38\(^{38,47,48}\) | 1.76            | 370             |
|       |                 |                 |                 |                 | 44.6            |                 |
| CdS   | 135\(^{38}\)   | 146             | 231             | 0.738           | 2.14            | 16              |
|       |                 |                 |                 |                 | 3.59            |                 |
| InSe  | 190\(^7\)     | 106             | 212             | 1.27            | 2.24            | 6.97            |
|       |                 |                 |                 |                 | 1.72            |                 |
| InN   | 660\(^{41,46}\)| 202             | 321             | 0.974\(^{46}\)  | 2.17            | 45              |
|       |                 |                 |                 |                 | 8.04            |                 |

FIG. 3. (a) Lattice thermal conductivity at 300K, (b) Debye temperature and (c) Grüneisen parameter of Wurzite structure semiconductors. The Debye temperatures plotted in (b) are \( \theta_a \), except for InSe and InN where \( \theta_D \) values are quoted in Refs. 7, 41, and 46.
TABLE IV. Lattice thermal conductivity at 300K, Debye temperature and Grüneisen parameter of rhombohedral semiconductors. The experimental Debye temperatures are \( \theta_D \) for Bi\(_2\)Te\(_3\) and Sb\(_2\)Te\(_3\), and \( \theta_c \) for Al\(_2\)O\(_3\). Units: \( \theta \) in (K), \( \kappa \) in (W/(m·K)).

| Comp. | \( \theta_D^{\exp} \) | \( \theta_D^{AGL} \) | \( \theta_c^{\exp} \) | \( \gamma^{\exp} \) | \( \gamma^{AGL} \) | \( \kappa^{\exp} \) | \( \kappa^{AGL} \) |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Bi\(_2\)Te\(_3\) | 155.5 | 155.7 | 155.7 | 155.7 | 155.7 | 155.7 | 155.7 |
| Sb\(_2\)Te\(_3\) | 160.7 | 160.7 | 160.7 | 160.7 | 160.7 | 160.7 | 160.7 |
| Al\(_2\)O\(_3\) | 390.87 | 390.87 | 390.87 | 390.87 | 390.87 | 390.87 | 390.87 |
| Cr\(_2\)O\(_3\) | 262.565 | 262.565 | 262.565 | 262.565 | 262.565 | 262.565 | 262.565 |
| Fe\(_2\)O\(_3\) | 182.388 | 182.388 | 182.388 | 182.388 | 182.388 | 182.388 | 182.388 |
| Bi\(_2\)Se\(_3\) | 104.177 | 104.177 | 104.177 | 104.177 | 104.177 | 104.177 | 104.177 |

\( \gamma \) were obtained using \( \gamma = 0.75 \) for BeO. Table 2.3 of Ref. 38 also cites an alternative value of \( \gamma = 1.38 \) for BeO (Table III). Using this outlier value would severely degrade the results down to 0.7, for the Pearson correlation, and 0.829, for the Spearman correlation. These values are too low for a reliable screening tool. This demonstrates the ability of the AGL calculations to compensate for anomalies in the experimental data when they exist and still provide a reliable screening method for the thermal conductivity.

**D. Rhombohedral materials**

Experimental results for rhombohedral materials (spacegroups: \( R\bar{3}mR \) (#166), \( R\bar{3}mH \) (#166) and \( R\bar{3}cH \) (#167); Pearson symbols: \( hR5 \), \( hR10 \)) are compared to the results of our calculations in Table IV and Figure 4. The experimental Debye temperatures are for \( \theta_D \) in the case of Bi\(_2\)Te\(_3\) and Sb\(_2\)Te\(_3\), and for \( \theta_c \) in the case of Al\(_2\)O\(_3\). The Pearson correlation between the experimental and calculated thermal conductivity values is 0.892. The corresponding Spearman correlation is 0.600. The Spearman correlation between the experimental values of the thermal conductivity and the values of \( \theta_c \) calculated with AGL is 0.943.

The thermal conductivity of Fe\(_2\)O\(_3\) is a clear outlier in this data set (see fig. 4). Its Grüneisen parameter, calculated with Equation (12), is 5.32. It is abnormally high. Equation (11) gives a similar value of 5.36, whereas Equation (16) gives a lower, but still very high, value of 4.06. Ignoring Fe\(_2\)O\(_3\) in the comparison increases the Pearson correlation of the calculated and experimental values of the thermal conductivity to 0.992, while the Spearman correlation increases to 0.9.

**E. Body-centred tetragonal materials**

Results for a set of body-centred tetragonal materials (spacegroup: \( I\bar{4}2d \) (#122); Pearson symbol: \( t\bar{1}6 \)) are shown in Table V and in Figure 5. For the materials ZnGeP\(_2\) and AgGaS\(_2\) there are three and two experimental values listed for \( \kappa^{\exp} \). This is due to the materials having different thermal conductivities in different crystalline directions\(^{52}\). The following results were obtained for the direction parallel to the optic axis, 36 W/(m·K) and 1.4 W/(m·K) for ZnGeP\(_2\) and AgGaS\(_2\), respectively. All of the experimental Debye temperatures listed in the table are the traditional Debye temperatures, \( \theta_D \).

The Pearson correlation between the AGL thermal conductivity values and the experimental values is 0.383. The corresponding Spearman correlation is 0.498. The Spearman correlation between the experimental values of the thermal conductivity and the calculated values of \( \theta_c \) is 0.401. The low correlations for this set of materials are due to their anisotropic structure, where the materials display different thermal conductivities along different lattice directions. This demonstrates the limits of the isotropic approximation made in the GIBBS method.

**FIG. 4.** Lattice thermal conductivity of rhombohedral semiconductors at 300K.

**TABLE V.** Lattice thermal conductivity at 300K, Debye temperature and Grüneisen parameter of body-centred tetragonal semiconductors. Units: \( \theta \) in (K), \( \kappa \) in (W/(m·K)).

| Comp. | \( \theta_D^{\exp} \) | \( \theta_D^{AGL} \) | \( \theta_c^{\exp} \) | \( \gamma^{\exp} \) | \( \gamma^{AGL} \) | \( \kappa^{\exp} \) | \( \kappa^{AGL} \) |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| CuGaTe\(_2\) | 226.7 | 226.7 | 226.7 | 226.7 | 226.7 | 226.7 | 226.7 |
| ZnGeP\(_2\) | 500 | 500 | 500 | 500 | 500 | 500 | 500 |
| ZnSiAs\(_2\) | 347 | 347 | 347 | 347 | 347 | 347 | 347 |
| CuInTe\(_2\) | 185 | 185 | 185 | 185 | 185 | 185 | 185 |
| AgGaS\(_2\) | 255 | 255 | 255 | 255 | 255 | 255 | 255 |
| CdGeP\(_2\) | 340 | 340 | 340 | 340 | 340 | 340 | 340 |
| CdGeAs\(_2\) | 140 | 140 | 140 | 140 | 140 | 140 | 140 |
| CuGaS\(_2\) | 350 | 350 | 350 | 350 | 350 | 350 | 350 |
| CuGaSe\(_2\) | 262 | 262 | 262 | 262 | 262 | 262 | 262 |
| ZnGeAs\(_2\) | 147 | 147 | 147 | 147 | 147 | 147 | 147 |
The results for materials with various other structures are shown in Table VI. The materials are CoSb_3 and IrSb_3 (spacegroup: Im3m (#204); Pearson symbol: cI32), ZnSb (spacegroup: Pbca (#61); Pearson symbol: aP16), Sb_2O_3 (spacegroup: Pnma (#56); Pearson symbol: aP20), InTe (spacegroup: Pm3m (#221); Pearson symbol: cP2), Bi_2O_3 (spacegroup: P121/c1 (#14); Pearson symbol: mP20), and SnO_2 (spacegroup: P4_2/mnm (#136); Pearson symbol: tP6). The experimental Debye temperatures listed in the table are the traditional Debye temperatures, $\theta_D$, with the exception of ZnSb for which it is $\theta_a$. For these materials, the Pearson correlation between the calculated and experimental values of the thermal conductivity is 0.914. The corresponding Spearman correlation is 0.071. The Spearman correlation between the experimental values of the thermal conductivity and the calculated values of $\theta_a$ is 0.143.

The low correlation values, particularly for the Spearman correlation, for this set of materials demonstrates the importance of the information about the material structure as an input for the AGL method. This is partly due to the fact that the Grüneisen parameter tends not to vary significantly between materials with a particular structure, thus reducing its effect on the ordinal ranking of the thermal conductivity of materials with the same structure.

### G. Half-Heusler materials

Carrete et al.\textsuperscript{2,61} studied the thermal conductivity of 107 half-Heusler (spacegroup: F43m (#216); Pearson symbol: cF12) compounds with \textit{ab initio} and machine learning techniques. In this section we compare their results with our AGL calculations. We first consider a subset of these half-Heusler materials, taken from Table I of Ref. 2, for which the thermal conductivity values were calculated using full anharmonic phonon parameterization solutions of the BTE. The thermal conductivities at 300K for this set of materials as calculated with Eq. (19) are shown in Table VII and in Figure 6. The Pearson correlation between the AGL thermal conductivity values and the full anharmonic phonon calculations is 0.495. The corresponding Spearman correlation is 0.810. The Spearman correlation between the full anharmonic phonon calculation values of the thermal conductivity and the values of $\theta_a$ as calculated with AGL is 0.730. A major contributor to the low Pearson correlation is the outlier calculated value of the thermal conductivity of FeNbP and NiPbTi, 109.0 W/(m·K). If these materials are removed from the dataset, the Pearson correlation increases to 0.629.

The second subset of half-Heusler materials studied is taken from Table III of Ref. 2, where the thermal conductivity was estimated using a machine learning algorithm. Comparison of these values with the thermal conductivity at 300K calculated with Eq. (19) is shown in Table VIII and Figure 7. The Pearson correlation between the AGL thermal conductivities and those produced by the machine learning algorithm is 0.578. The corresponding
TABLE VII. Thermal conductivities of half-Heusler semiconductors at 300K compared to full anharmonic phonon ab-initio parameterization from Ref. 2. Units: $\theta$ in (K), $\kappa$ in (W/(m·K)).

| Comp.          | $\theta^{\text{AGL}}$ | $\theta^{\text{ML}}$ | $\gamma^{\text{ML}}$ | $\kappa^{\text{anh}}$ | $\kappa^{\text{ML}}$ |
|----------------|------------------------|------------------------|------------------------|------------------------|------------------------|
| AgKTe          | 105                    | 152                    | 2.26                   | 0.508                  | 1.0                    |
| BeNaP          | 302                    | 436                    | 2.05                   | 4.08                   | 5.3                    |
| BiBaK          | 95                     | 137                    | 1.94                   | 2.19                   | 1.59                   |
| BiKrSr         | 99                     | 143                    | 1.96                   | 1.96                   | 1.45                   |
| BiLiSr         | 126                    | 182                    | 1.94                   | 3.04                   | 2.48                   |
| CoAsZr         | 306                    | 442                    | 2.14                   | 24.0                   | 17.51                  |
| CoBiHf         | 204                    | 294                    | 2.17                   | 18.6                   | 10.43                  |
| CoSbZr         | 231                    | 333                    | 3.00                   | 25.0                   | 4.69                   |
| CoScSe         | 230                    | 331                    | 2.09                   | 15.0                   | 6.64                   |
| CoSiTa         | 296                    | 427                    | 1.92                   | 37.8                   | 23.06                  |
| FeNbP          | 343                    | 495                    | 1.94                   | 109.0                  | 23.79                  |
| GeCaZn         | 197                    | 284                    | 2.05                   | 2.75                   | 4.53                   |
| GeNaY          | 189                    | 273                    | 2.04                   | 8.06                   | 4.28                   |
| LiBaSr         | 88                     | 127                    | 1.33                   | 0.582                  | 1.84                   |
| IrPTi          | 309                    | 446                    | 2.18                   | 27.4                   | 20.25                  |
| NiPbTi         | 205                    | 296                    | 2.20                   | 109.0                  | 23.79                  |
| NiSbSc         | 232                    | 334                    | 2.03                   | 19.5                   | 9.13                   |
| NiSnTi         | 249                    | 359                    | 2.06                   | 17.9                   | 10.7                   |
| NiSnZr         | 229                    | 330                    | 2.06                   | 19.6                   | 10.22                  |
| OsSiTa         | 227                    | 328                    | 2.14                   | 29.6                   | 16.62                  |
| PdAsY          | 230                    | 332                    | 2.17                   | 5.48                   | 9.43                   |
| PdSrTe         | 130                    | 188                    | 2.13                   | 1.16                   | 2.44                   |
| PtGaTa         | 242                    | 349                    | 2.19                   | 32.9                   | 16.78                  |
| PtGeTi         | 263                    | 379                    | 2.23                   | 16.9                   | 14.41                  |
| PtLaNb         | 140                    | 202                    | 2.69                   | 16.5                   | 2.2                    |
| RhHSb          | 232                    | 335                    | 2.18                   | 21.8                   | 14.06                  |
| RhNbSi         | 345                    | 497                    | 2.09                   | 15.3                   | 26.15                  |
| RuAsV          | 334                    | 482                    | 2.19                   | 23.5                   | 21.37                  |
| SbCaK          | 141                    | 203                    | 1.92                   | 2.70                   | 2.47                   |
| SiCdSr         | 168                    | 242                    | 2.05                   | 13.5                   | 3.79                   |
| SnBaSr         | 114                    | 165                    | 1.71                   | 2.01                   | 3.19                   |
| TeAgLi         | 166                    | 239                    | 2.32                   | 1.52                   | 2.79                   |

Spearman correlation is 0.706. The Spearman correlation between the machine learning thermal conductivities and the AGL values of $\theta_\alpha$ is 0.679.

Experimental results for the thermal conductivity of 7 of these half-Heusler materials were available in the literature, and these values are shown in Table IX and Figure 8. The Pearson correlation between the AGL thermal conductivities and the experimental values is 0.064, while the corresponding Spearman correlation is $-0.036$. The Spearman correlation between the experimental thermal conductivities and the AGL values of $\theta_\alpha$ is 0.0. However, this is a small sample set, and these low correlation values appear to be primarily due to the outlier material CoSbZr, for which AGL predicts a relatively high value of 3.0 for the Grüneisen parameter. Ignoring this material in the comparison increases the Pearson correlation between the thermal conductivities to 0.262 and the Spearman correlation to 0.314.

H. AGL predictions for zincblende materials

In order to demonstrate the potential utility of the AGL method for high-throughput screening of the thermal properties of materials we have calculated the Debye temperature, Grüneisen parameter and thermal conductivity for 45 zincblende structure (spacegroup: $F\overline{4}3m$ (#216); Pearson symbol: cF8) materials which were not included in Table I, and for which experimental values of the thermal conductivity do not seem to be available in the literature. The results for these materials are shown in Table X and in figure 9.

From these results, it is noticeable that BeO is predicted to have the highest thermal conductivity, with a value similar to that of SiC. This high thermal conductivity is in agreement with recent first principles calculations. Another set of materials predicted to have high thermal conductivity includes the nitrides PrN, ReN, NbN and...
TABLE VIII. Thermal conductivities of half-Heusler semiconductors at 300K compared to machine learning algorithm predictions from Ref. 2. Units: $\theta$ in (K), $\kappa$ in (W/(m·K)).

| Comp.  | $\theta_{AGL}$ | $\gamma_{AGL}$ | $\kappa_{ML}[2]$ | $\kappa_{AGL}$ |
|--------|----------------|----------------|------------------|----------------|
| AuAlHf | 217            | 313            | 2.12             | 16.7           |
| BLiSi  | 433            | 624            | 2.07             | 62.1           |
| BiBaK  | 95             | 137            | 1.94             | 1.24           |
| CoAsHf | 266            | 383            | 2.13             | 20.0           |
| CoAsTi | 345            | 497            | 2.15             | 37.1           |
| CoAsZr | 306            | 442            | 2.14             | 27.7           |
| CoBiHf | 204            | 294            | 2.17             | 22.5           |
| CoBiTi | 236            | 341            | 2.19             | 27.1           |
| CoBiZr | 223            | 322            | 2.17             | 17.8           |
| CoGeNh | 295            | 425            | 2.39             | 36.2           |
| CoGeTa | 266            | 383            | 2.24             | 27.2           |
| CoGeV  | 334            | 482            | 2.01             | 29.1           |
| CoHfSb | 190            | 274            | 1.69             | 21.9           |
| CoNiSi | 323            | 466            | 2.18             | 30.1           |
| CoNiSn | 238            | 343            | 2.56             | 20.7           |
| CoSbTi | 263            | 379            | 2.10             | 23.3           |
| CoSbZr | 231            | 333            | 3.0              | 24.4           |
| CoSiTa | 296            | 427            | 1.92             | 36.9           |
| CoSnTa | 217            | 313            | 3.32             | 22.7           |
| CoSnV  | 266            | 383            | 2.49             | 19.8           |
| FeAsNb | 339            | 489            | 2.13             | 47.6           |
| FeAsTa | 295            | 425            | 2.13             | 32.9           |
| FeGeV  | 245            | 354            | 1.40             | 32.8           |
| FeHfSb | 216            | 311            | 1.70             | 29.1           |
| FeSbTa | 196            | 282            | 1.65             | 31.2           |
| FeSbV  | 305            | 440            | 1.50             | 24.1           |
| FeTeTi | 266            | 384            | 2.18             | 26.2           |
| GeAlLi | 270            | 390            | 2.06             | 16.5           |
| GeAsTi | 277            | 399            | 2.22             | 30.1           |
| GeAsZr | 255            | 368            | 2.19             | 17.4           |
| GeBiZr | 206            | 297            | 2.24             | 12.8           |
| GeGeNb | 279            | 402            | 2.17             | 33.0           |
| GeGeTa | 256            | 369            | 2.15             | 37.2           |
| GeSbV  | 288            | 416            | 2.19             | 30.0           |
| GeHfSb | 221            | 319            | 2.20             | 24.7           |
| GeSnTa | 232            | 334            | 2.18             | 19.8           |
| GeSnZr | 218            | 314            | 2.23             | 22.1           |
| NiAsSc | 300            | 432            | 2.11             | 17.5           |

TABLE IX. Thermal conductivities of half-Heusler semiconductors at 300K compared to experimental values. Units: $\theta$ in (K), $\kappa$ in (W/(m·K)).

| Comp.  | $\theta_{AGL}$ | $\gamma_{AGL}$ | $\kappa_{ML}[2]$ | $\kappa_{AGL}$ | $\kappa_{exp}$ |
|--------|----------------|----------------|------------------|----------------|----------------|
| CoHfSb | 190            | 274            | 1.69             | 21.9           | 21.8           |
| CoSbTa | 263            | 379            | 2.10             | 23.3           | 23.1           |
| CoSbZr | 231            | 333            | 3.0              | 24.4           | 24.3           |
| FeSbV  | 230            | 332            | 2.08             | 19.5           | 19.5           |
| FeSnTa | 249            | 359            | 2.06             | 16.8           | 16.8           |
| NiAsSc | 300            | 432            | 2.11             | 17.5           | 17.5           |

MoN. Although BAs was previously predicted to have an extremely high thermal conductivity, the AGL value is only slightly higher than that of Si or AlP, and less than that of BP, BN or SiC. The materials with the lowest thermal conductivity in this set are AgI and CuI. AgI, in particular, is predicted by AGL to have a thermal conductivity lower than that of any of the materials in Table I.
TABLE X. Lattice thermal conductivity at 300K, Debye temperature and Grüneisen parameter of Zincblende structure materials for which the experimental Debye temperature is not available in the literature. Units: $\theta$ in (K), $\kappa$ in (W/(m·K)).

| Comp. | $\theta_{\text{AGL}}$ | $\theta_{\text{exp}}$ | $\gamma_{\text{AGL}}$ | $\kappa_{\text{AGL}}$ |
|-------|-------------------|-------------------|-------------------|-------------------|
| AgI   | 123               | 155               | 2.45              | 1.51              |
| AgO   | 247               | 311               | 2.06              | 7.19              |
| AgSe  | 225               | 255               | 2.52              | 3.09              |
| AuN   | 259               | 326               | 2.49              | 9.03              |
| BaS   | 420               | 529               | 1.95              | 25.75             |
| BeO   | 845               | 1065              | 1.74              | 62.77             |
| BeSe  | 324               | 408               | 1.80              | 15.6              |
| CaSe  | 208               | 262               | 1.84              | 6.78              |
| CdS   | 228               | 287               | 2.15              | 6.91              |
| CoO   | 427               | 538               | 2.41              | 14.46             |
| CuBr  | 190               | 239               | 2.44              | 2.94              |
| CuCl  | 243               | 295               | 2.40              | 3.76              |
| CuF   | 272               | 343               | 2.34              | 4.66              |
| CuI   | 161               | 203               | 2.48              | 2.45              |
| GaBi  | 140               | 177               | 2.18              | 3.32              |
| GeO   | 375               | 450               | 2.97              | 4.90              |
| GeP   | 239               | 301               | 1.54              | 11.5              |
| GeSe  | 231               | 291               | 1.78              | 4.01              |
| HfN   | 348               | 439               | 1.89              | 36.14             |
| HgSe  | 176               | 222               | 2.34              | 4.35              |
| IrN   | 371               | 467               | 2.19              | 31.79             |

FIG. 9. Predicted thermal conductivities of zincblende structure materials at 300K. The AGL values for the materials with experimental data listed in Table I are shown in black (see also figure 1). AGL predictions for materials with no experimental data are in red.

IV. CONCLUSIONS

We implemented the “GIBBS” quasi-harmonic Debye model in the AGL software package within the AFLOW and Materials Project high-throughput computational materials science frameworks. We used it to automatically calculate the thermal conductivity, Debye temperature and Grüneisen coefficient of materials with various structures and compared them with experimental results. A major aim of high-throughput calculations is to identify useful markers (descriptors) for screening large datasets of structures for desirable properties. In this study we examined whether the inexpensive-to-calculate Debye model thermal properties may be useful as such markers for high thermal conductivity materials, despite the well known deficiencies of this model in their quantitative evaluation. We therefore concentrated on correlations between the calculated quantities and the corresponding experimental data.

The correlations between the experimental values of the thermal conductivity and those calculated with AGL are summarized in Table XI. For the entire set of materials examined we find a high Pearson correlation of 0.879 between $\kappa_{\text{exp}}$ and $\kappa_{\text{AGL}}$. It is particularly high, above 0.9, for materials with high symmetry (cubic or rhombohedral) structures, but significantly lower for anisotropic materials. We also compared these results with similar calculations of the thermal conductivity, using the experimental values of the Debye temperature and Grüneisen coefficient. The two methods gave similar Pearson correlations for the thermal conductivities, demonstrating that the AGL approach can rectify the lack of this experimental data in screening large data sets of materials.

The Spearman correlation between $\kappa_{\text{exp}}$ and $\theta_{\text{AGL}}$ for the entire set of materials is almost as high as the Pearson correlation between the calculated and experimental conductivities. It is, however, less consistent for the high symmetry structures, with a relatively low value of 0.645 for the Rocksalt structures. The Spearman correlation between $\kappa_{\text{exp}}$ and $\kappa_{\text{AGL}}$ is found to be inferior to both previous measures as a descriptor of high conductivity materials. The correlations for the half-Heusler materials are summarized in Table XII.

Overall, despite the quantitative limitations of the
method, the AGL approach can be useful for quickly screening large data sets of materials for favorable thermal properties.

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* On leave from the Department of Physics, NRCN, Israel steffano@duke.edu
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