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An efficient and accurate framework for calculating lattice thermal conductivity of solids: AFLOW—AAPL Automatic Anharmonic Phonon Library

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One of the most accurate approaches for calculating lattice thermal conductivity, κ_{ℓ} , is solving the Boltzmann transport equation starting from third-order anharmonic force constants. In addition to the underlying approximations of ab-initio parameterization, two main challenges are associated with this path: high computational costs and lack of automation in the frameworks using this methodology, which affect the discovery rate of novel materials with ad-hoc properties. Here, the Automatic Anharmonic Phonon Library (AAPL) is presented. It efficiently computes interatomic force constants by making effective use of crystal symmetry analysis, it solves the Boltzmann transport equation to obtain κ_{ℓ} , and allows a fully integrated operation with minimum user intervention, a rational addition to the current high-throughput accelerated materials development framework AFLOW. An “experiment vs. theory” study of the approach is shown, comparing accuracy and speed with respect to other available packages, and for materials characterized by strong electron localization and correlation. Combining AAPL with the pseudo-hybrid functional ACBN0 is possible to improve accuracy without increasing computational requirements.

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INTRODUCTION

Lattice thermal conductivity, κ_{ℓ} , is the key materials' property for many technologies and applications, such as thermoelectric materials,^{1–3} heat sink materials,⁴ rewritable density scanning-probe phase-change memories,⁵ and thermal medical devices.⁶ Fast and robust predictions of this quantity remain a challenge:⁷ semi-empirical models^{8–10} are computationally inexpensive but require some experimental data. Similarly, classical molecular dynamics combined with Green–Kubo relations^{11–13} is reasonably quick but requires the knowledge of specific force fields. On the contrary, frameworks based on the quasiharmonic Debye model, such as GIBBS¹⁴ or the Automatic-Gibbs-Library (AGL),^{15,16} are extremely efficient as pre-screening techniques but they lack quantitative accuracy.

The quasiharmonic approximation (QHA), alone has also been used in different models to predict κ_{ℓ} .^{7,17} Although QHA-based models overall improve accuracy of κ_{ℓ} , they are far from the results obtained from calculating the anharmonic force constants and solving the associated Boltzmann transport equation (BTE).^{8,18} To the best of our knowledge, solving the BTE is the optimal method for systematically and accurately calculating thermal conductivity.^{19–21} This approach has been successfully applied to many systems during the last decade. It has been recently implemented in packages including Phono3py,²² PhonTS,²³ ALAMODE,²⁴ and ShengBTE,²⁵ which compute κ_{ℓ} by calculating the anharmonic force constants and solving the BTE. Nevertheless,

there is a lack of a robust framework able to calculate κ_{ℓ} with minimum intervention from the user, and therefore targeted to high-throughput automated and accelerated materials discovery.

Many challenges need to be tackled. **I.** The third-order interatomic force constants (IFCs) up to a certain distance cut-off are computationally expensive to obtain from first principles. Overall they represent the major concern for the method. Effective use of crystalline symmetry of the system must be employed to map, through appropriate tensorial transformations, dependent IFCs and therefore reduce the number of calculations. The task is performed by the internal AFLOW point-factor-space group calculator.²⁶ Recently, it has also been proposed to obtain the IFCs by inverting the results of many entangled calculations with the use of compressive sensing.²⁷ Further studies need to be carried out to address the scaling of the algorithm with respect to cut-offs and accuracy. **II.** For a rational software for accelerated materials development, all the geometric optimizations, symmetry analyses, supercell creation, pre-processing and post-processing, and automatic error corrections to get the IFCs, in addition to the appropriate integration for the BTE must be performed by a single code. Here, we present Automatic Anharmonic Phonon Library (AAPL), which computes the IFCs and solves the BTE to predict κ_{ℓ} as part of the AFLOW high-throughput framework,^{28–39} automating the entire process. The software is being finalized for an official open-source release during 2017, within the GNU GPL license. **III.** The accuracy of the method mostly depends on the

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accuracy of the computed forces, and therefore it will inherit the same limitations as the ab-initio method used. For materials characterized by strong electron localization and correlation, accurate hybrid functionals for Density Functional Theory parameterizations might not even be feasible, as they would drastically increase computational costs, with respect to more basic LDA or GGA functionals. In that case, new strategies should be developed to contain computational demands. Here, an example is given: combining AAPL with the pseudo-hybrid functional Agapito Curtarolo Buongiorno Nardelli ab-initio DFT functional (ACBN0) improves the accuracy without increasing computational requirements.^{40–46}

RESULTS AND DISCUSSION

The Automatic Anharmonic Phonon Library

The Boltzmann transport equation. The Boltzmann equation for phonons, originally formulated by Peierls in 1929, is an important approach for studying phonon transport.⁸ Its solution has posed a challenge for the last several decades. Callaway⁹ and Allen⁴⁷ proposed models based on parameters that are fitted to experimental data. In 2003, Deinzer et al. used density functional perturbation theory (DFPT) to study the phonon linewidths of Si and Ge.⁴⁸ Since then, many authors have used the solution of the BTE to calculate the lattice thermal conductivity of solids.^{19–21} The most used approach is the iterative solution of the BTE proposed by Omini et al. and successfully applied in the prediction of the κ_ℓ tensor for different materials.^{49–51}

$$\kappa_\ell^{\alpha\beta} = \frac{1}{N\Omega k_B T^2} \sum_\lambda f_0(f_0 + 1) (\hbar\omega_\lambda)^2 v_\lambda^\alpha F_\lambda^\beta, \quad (1)$$

where superscripts α and β are two of the Cartesian direction indices and the subscript λ comprises both phonon branch index i and a wave vector $\mathbf{q}$. The variables ω_λ and $\mathbf{v}_\lambda$ are the angular frequency and group velocity of the phonon mode λ , respectively, while $f_0(\omega_\lambda)$ is the phonon distribution function according to Bose–Einstein statistics. All these quantities are obtained through the calculation of the IFCs by using a finite-difference supercell approach: forces vs. small displacement of inequivalent atoms. In this approach, a reference unit cell of volume Ω is used to create the supercell up to the cut-off distance. For the various summations, the Brillouin zone, BZ, is discretized into a Γ -centered orthogonal regular grid of $N \equiv N_1 \times N_2 \times N_3$ $\mathbf{q}$ -points, where subscripts 1, 2, and 3 indicate the lattice vector indices.

The mean-free displacement $\mathbf{F}_\lambda$ follows the definition of the Bose–Einstein phonon distribution, f_λ , in the presence of a temperature gradient ∇T . For small perturbations, $\nabla T \sim 0$, f_λ can be expanded as $f_\lambda \sim f_0(\omega_\lambda) + g_\lambda$, where g_λ is the first-order non-equilibrium contribution linear in ∇T :

$$g_\lambda \equiv -\mathbf{F}_\lambda \cdot \nabla T \frac{df_0}{dT}.$$

Finally, the BTE can be expressed as a linear system of equations for $\mathbf{F}_\lambda$, as^{20,49–53}

$$\begin{aligned} \mathbf{F}_\lambda &= \tau_\lambda^0 (\mathbf{v}_\lambda + \mathbf{\Delta}_\lambda) \\ \mathbf{\Delta}_\lambda &= \frac{1}{N} \left(\sum_{\lambda'\lambda''}^+ \Gamma_{\lambda\lambda'\lambda''}^+ (\xi_{\lambda\lambda'} \mathbf{F}_{\lambda''} - \xi_{\lambda\lambda''} \mathbf{F}_{\lambda'}) \right. \\ &\quad + \sum_{\lambda'\lambda''}^- \frac{1}{2} \Gamma_{\lambda\lambda'\lambda''}^- (\xi_{\lambda\lambda'} \mathbf{F}_{\lambda''} + \xi_{\lambda\lambda''} \mathbf{F}_{\lambda'}) \\ &\quad \left. + \sum_{\lambda'} \Gamma_{\lambda\lambda'} \xi_{\lambda\lambda'} \mathbf{F}_{\lambda'} \right), \end{aligned} \quad (2)$$

with $\xi_{\lambda\lambda'} = \omega_\lambda/\omega_{\lambda'}$. The frequently used relaxation time approximation (RTA), corresponds to neglecting the $\mathbf{\Delta}_\lambda$ correction. For the full solution, $\mathbf{F}_\lambda$ can be self-consistently solved starting from the RTA guess, until convergence of κ_ℓ , Eq. (1). The other quantities present

in these formulas, the relaxation time τ_λ^0 , and the three-phonon scattering rates $\Gamma_{\lambda\lambda'\lambda''}^\pm$, will be illustrated in the next section.

Vibrational modes and group velocities. The vibrational modes are obtained by diagonalizing the dynamical matrix $D(\mathbf{q})$ ^{8,54–56}

$$D(\mathbf{q})\mathbf{e}_\lambda = \omega_\lambda^2 \mathbf{e}_\lambda, \quad (3)$$

$$D_{ij}^{\alpha\beta}(\mathbf{q}) = \sum_l \frac{\Phi(i, j)_{\alpha\beta}}{\sqrt{M(i)M(j)}} \exp[-i\mathbf{q} \cdot (\mathbf{R}_l - \mathbf{R}_0)], \quad (4)$$

where $M(j)$ is the mass of j atom, $\mathbf{e}_\lambda$ is the eigenvector for λ , $\mathbf{R}_l$ is the position of lattice point l and $\Phi(i, j)_{\alpha\beta}$ are the second-order force constants. $D(\mathbf{q})$ is a Hermitian $3n_a \times 3n_a$ matrix, where the factor “3” comes from the dimensionality of the problem, and n_a represents the number of atoms in the unit cell.

The non-analytical contributions to the dynamical matrix are included by using the mixed-space formulation of Wang et al.⁵⁷

$$\tilde{D}_{ij}^{\alpha\beta}(\mathbf{q}) = \frac{4\pi e^2}{\Omega} \frac{[\mathbf{q} \cdot \mathbf{Z}(i)]_\alpha [\mathbf{q} \cdot \mathbf{Z}(j)]_\beta}{\mathbf{q} \cdot \epsilon_\infty \cdot \mathbf{q}} \exp[-i\mathbf{q} \cdot (\mathbf{R}_l - \mathbf{R}_0)]. \quad (5)$$

This contribution requires the calculation of the Born effective charge tensors, $\mathbf{Z}$, and the high-frequency static dielectric tensor, ϵ_∞ , i.e. the contribution to the dielectric permittivity tensor from the electronic polarization.⁵⁸ The mixed-space approach has been successfully applied in the calculation of phonon and thermal properties for a wide range of polar materials.⁵⁹ Materials with high $\mathbf{Z}$ and low ϵ_∞ are the cases in which the non-analytical contributions are crucial for appropriate description of the phonon spectra as they cause the LO–TO splitting of the spectrum (between longitudinal and transverse optical phonon frequencies).⁵⁸

The group velocities, $\mathbf{v}_\lambda$, follow the Hellmann–Feynman theorem:

$$\mathbf{v}_\lambda = \frac{1}{2\omega_\lambda} \left\langle \mathbf{e}_\lambda \left| \frac{\partial D(\mathbf{q})}{\partial \mathbf{q}} \right| \mathbf{e}_\lambda \right\rangle. \quad (6)$$

Scattering time. The total scattering time is a sum of terms representing different phenomena:

$$\frac{1}{\tau_\lambda^0} = \frac{1}{\tau_\lambda^{\text{anh}}} + \frac{1}{\tau_\lambda^{\text{iso}}} + \frac{1}{\tau_\lambda^{\text{bnd}}}. \quad (7)$$

$\tau_\lambda^{\text{iso}}$ indicates the isotopic or elastic scattering time and it is due to the isotopic disorder^{60,61}

$$\begin{aligned} \frac{1}{\tau_\lambda^{\text{iso}}} &= \frac{1}{N} \sum_{\lambda'} \Gamma_{\lambda\lambda'} \\ &= \frac{1}{N} \sum_{\lambda'} \frac{\pi\omega_\lambda^2}{2} \sum_i g(i) |\mathbf{e}_\lambda^*(i) \mathbf{e}_{\lambda'}(i)|^2 \delta(\omega_\lambda - \omega_{\lambda'}), \end{aligned} \quad (8)$$

where $g(i) = \sum_s f_s(i) [1 - M(i)^s / \bar{M}(i)^s]^2$ is the Pearson deviation coefficient of masses $M(i)^s$ of isotopes s for the i atom, f_s is the relative fraction of isotope s , and $\bar{M}(i)^s$ is the average mass of the element.⁶²

$\tau_\lambda^{\text{bnd}}$ is the time associated with scattering at the grain boundaries^{63,64}

$$\frac{1}{\tau_\lambda^{\text{bnd}}} = \frac{|\mathbf{v}_\lambda|}{L}, \quad (9)$$

where L is the average grain size. The effect of the boundaries on κ_ℓ can also be calculated by restricting the summation to the modes with a mean free path, $\Lambda = \mathbf{F}_\lambda \cdot \mathbf{v}_\lambda / |\mathbf{v}_\lambda|$, shorter than L .²⁵

$$\kappa_{\ell, (\Lambda < L)}^{\alpha\beta} = \frac{1}{N\Omega k_B T^2} \sum_{\lambda}^{\Lambda < L} f_0(f_0 + 1) (\hbar\omega_\lambda)^2 v_\lambda^\alpha F_\lambda^\beta. \quad (10)$$

$\tau_{\lambda}^{\text{anh}}$ is the three-phonon scattering time. It is the largest contribution to τ_{λ}^0 for single crystals at medium-temperature ranges and it is the most computationally expensive quantity to obtain:

$$\frac{1}{\tau_{\lambda}^{\text{anh}}} = \frac{1}{N} \left(\sum_{\lambda\lambda''}^{+} \Gamma_{\lambda\lambda''}^{+} + \sum_{\lambda\lambda''}^{-} \frac{1}{2} \Gamma_{\lambda\lambda''}^{-} \right). \quad (11)$$

Conservation of the quasi-momentum requires that $\mathbf{q}'' = \mathbf{q} \pm \mathbf{q}' + \mathbf{Q}$ in the summation $\Sigma^{\pm}$, for some reciprocal lattice vector $\mathbf{Q}$ such that $\mathbf{q}''$ is in the same image of the Brillouin zone as $\mathbf{q}$ and $\mathbf{q}'$. The three-phonon scattering rates, $\Gamma_{\lambda\lambda''}^{\pm}$, are computed as

$$\Gamma_{\lambda\lambda''}^{+} \equiv \frac{\hbar\pi}{4} \frac{f_0' - f_0''}{\omega_{\lambda}\omega_{\lambda'}\omega_{\lambda''}} |V_{\lambda\lambda''}^{+}|^2 \delta(\omega_{\lambda} + \omega_{\lambda'} - \omega_{\lambda''}), \quad (12)$$

and

$$\Gamma_{\lambda\lambda''}^{-} \equiv \frac{\hbar\pi}{4} \frac{f_0' + f_0'' + 1}{\omega_{\lambda}\omega_{\lambda'}\omega_{\lambda''}} |V_{\lambda\lambda''}^{-}|^2 \delta(\omega_{\lambda} - \omega_{\lambda'} - \omega_{\lambda''}). \quad (13)$$

The scattering matrix elements, $V_{\lambda\lambda''}^{\pm}$, are given by^{52,53}

$$V_{\lambda\lambda''}^{\pm} = \sum_{\substack{\{i,j,k\} \in \text{uc} \\ \{j,k\} \in \text{sc} \\ \alpha\beta\gamma}} \Phi(i,j,k)_{\alpha\beta\gamma} \frac{e_{p',\pm\mathbf{q}'}^{\alpha}(i) e_{p'',-\mathbf{q}''}^{\beta}(j) e_{p,\mathbf{q}}^{\gamma}(k)}{\sqrt{M(i)M(j)M(k)}}, \quad (14)$$

where $\Phi(i,j,k)_{\alpha\beta\gamma}$ are the anharmonic force constants (introduced below) and $e_{p',\pm\mathbf{q}'}^{\alpha}(j)$ is the element of the eigenvector of branch p' at point $\pm\mathbf{q}'$ that corresponds to j atom in the β -direction. Note the indices $\{i \in \text{uc} (\text{unit cell})\}$ while $\{j, k \in \text{sc} (\text{supercell})\}$. The conservation of energy, enforced by the Dirac distribution, can cause numerical instability during the calculations. Thus, we follow Li et al.²⁵ and substitute δ with a normalized Gaussian distribution $g: \delta(\dots) \rightarrow g(\dots)$ in Eqs. (12) and (13) with

$$\begin{aligned} g(\omega_{\lambda} - (\pm\omega_{\lambda'} + \omega_{\lambda''})) &\equiv \frac{1}{\sqrt{2\pi\sigma}} e^{-\frac{(\omega_{\lambda} - (\pm\omega_{\lambda'} + \omega_{\lambda''}))^2}{2\sigma^2}}, \\ \sigma &\equiv \zeta \sigma_{(\pm\omega_{\lambda'} + \omega_{\lambda''})} \\ &= \frac{\zeta}{\sqrt{12}} \sqrt{\sum_{\nu} \left[\sum_{\alpha} (v_{\lambda'}^{\alpha} - v_{\lambda''}^{\alpha}) \frac{Q_{\nu}^{\alpha}}{N_{\nu}} \right]^2}, \end{aligned} \quad (15)$$

where Q_{ν}^{α} is the component in the Cartesian direction, α , of the reciprocal-space lattice vector $\mathbf{Q}_{\nu}$ and N_{ν} is the number of points of the $\mathbf{q}$ -points grid in the reciprocal-space direction ν . In principle, the parameter ζ could be taken equal to one. However, it can be adjusted to lower values to increase the speed of the calculations, without much effect on the overall accuracy of the integrations.

Interatomic force constants. The n^{th} -order IFCs, $\Phi(i,j,\dots)_{\alpha\beta\dots}$ are tensorial quantities representing derivatives of the potential energy (V) with respect to the atomic displacements from equilibrium:

$$\begin{aligned} V &= V_0 + \frac{1}{2!} \sum_{ij,\alpha\beta} \Phi(i,j)_{\alpha\beta} r(i)^{\alpha} r(j)^{\beta} \\ &+ \frac{1}{3!} \sum_{ijk,\alpha\beta\gamma} \Phi(i,j,k)_{\alpha\beta\gamma} r(i)^{\alpha} r(j)^{\beta} r(k)^{\gamma} + \dots \end{aligned} \quad (16)$$

Labels $i, j, k, \dots$ span atoms of the cell and indices $\alpha, \beta, \gamma, \dots$ are the Cartesian directions of the displacement. Second-order harmonic IFC, $\Phi(i,j)_{\alpha\beta}$, calculations were already implemented in the original harmonic Automatic-Phonon-Library (APL),²⁸ which obtains dispersion curves using three different approaches: direct force constant,^{65–67} linear response and projector-augmented wave (PAW) potentials,⁶⁸ and the frozen phonon methods.^{69,70}

Third-order IFCs, $\Phi(i,j,k)_{\alpha\beta\gamma}$ contain information about the anharmonicity of the lattice and they tend to rule phonon scattering in single crystals in the medium-temperature ranges.^{71,72} Given the choice of a supercell size, the finite

difference method to calculate the third-order IFCs leads to:

$$\begin{aligned} \Phi(i,j,k)_{\alpha\beta\gamma} &\equiv \frac{\partial^3 V}{\partial r(i)^{\alpha} \partial r(j)^{\beta} \partial r(k)^{\gamma}} \\ &\simeq \frac{1}{2h} \left[\frac{\partial^2 V}{\partial r(j)^{\beta} \partial r(k)^{\gamma}} (h(i)^{\alpha}) - \frac{\partial^2 V}{\partial h(j)^{\beta} \partial r(k)^{\gamma}} (-h(i)^{\alpha}) \right] \\ &\simeq \frac{1}{4h^2} \left[-\psi(h(i)^{\alpha}, h(j)^{\beta}, k)_{\gamma} + \psi(-h(i)^{\alpha}, h(j)^{\beta}, k)_{\gamma} \right. \\ &\quad \left. + \psi(h(i)^{\alpha}, -h(j)^{\beta}, k)_{\gamma} - \psi(-h(i)^{\alpha}, -h(j)^{\beta}, k)_{\gamma} \right] \end{aligned} \quad (17)$$

where $\{\pm h(i)^{\alpha}\}$, $\{\pm h(j)^{\beta}\}$ represent displacements of magnitude h of the i, j atoms in the Cartesian directions $\pm\alpha, \pm\beta$ and $\psi(\pm h(i)^{\alpha}, \pm h(j)^{\beta}, k)_{\gamma}$ are the γ components of the forces felt by the k atom in the distorted configurations caused by the i and j atoms.

The third-order IFCs' calculation is computationally intensive: each $\Phi(i,j,k)_{\alpha\beta\gamma}$ requires four supercell calculations (Eq. (17)). Effective use of crystal symmetry can help the process.⁷³ AAPL uses point, factor, and space group symmetry operations computed by the AFLOW symmetry engine²⁶ to identify equivalence between single, pairs, and triplets of atoms (positions) and test equivalence between other field quantities, such as differential Φ or finite difference forces ψ (covariantly transforming). To avoid confusion, here indices as superscripts $\{\alpha\beta\gamma\dots\}$ or subscripts $\{\alpha\beta\gamma\dots\}$ are used to identify the character of the symmetry transformation to be applied.⁷⁴

The reduction of third-order IFC calculations is performed through the following steps:

1. Inequivalent atoms, pairs, and triplets are identified using space group symmetries. The user chooses the neighbor-shell cut-off and only pairs/triplets completely contained are considered.
2. The IFC tensors belonging to inequivalent triplets are analyzed. The symmetry operations mapping the representative inequivalent to the equivalent Φ are saved: $\Phi(i,j,k)_{\alpha\beta\gamma} \rightarrow \Phi'(i',j',k')_{\alpha'\beta'\gamma'}$.
3. Each inequivalent tensor $\Phi(i,j,k)_{\alpha\beta\gamma}$ contains $3 \times 3 \times 3 = 27$ coefficients. Every static ab-initio calculation produces the vectorial force field for all the k atoms of the supercell (where each k , combined with the inequivalent pair (i,j) , possibly generates (i,j,k) inequivalent triplets) starting from a combination of deformed positions for the i and j atoms belonging to inequivalent pairs. This requires the evaluation of $3 \times 3 = 9$ configurations. Following Eq. (17) four forces $\psi(\pm h(i)^{\alpha}, \pm h(j)^{\beta}, k)_{\gamma}$ are required for every entry $\Phi(i,j,k)_{\alpha\beta\gamma}$. To conclude, a total of 36 static calculations are necessary to parameterize $\Phi(i,j, \forall k \in \text{sc})$.
4. A large look-up table of all the necessary finite difference forces $\psi(h(i)^{\alpha}, h(j)^{\beta}, k)$ is prepared at the beginning of the process. Every ψ can be constituent of many inequivalent $\Phi(i,j,k)_{\alpha\beta\gamma}$, and, within each Φ , be a term in several internal coefficients. To exploit redundancy, the force field generated by every static ab-initio calculation is mapped through symmetry operations to recover as many possible other $\psi(h(i)^{\alpha}, h(j)^{\beta}, \forall k \in \text{sc}) \rightarrow \psi(h(i')^{\alpha'}, h(j')^{\beta'}, \forall k' \in \text{sc})_{\gamma'}$. Calculated and symmetry reproduced ψ are then removed from the table, and the algorithm moves to the next one to characterize. The process is repeated until all the ψ are found. The process guarantees that only the minimum amount of calculations are performed, compatible with the model of Eq. (17).
5. During the process, many equivalent entries of the tensors $\Phi(i,j,k)_{\alpha\beta\gamma}$ are generated by the static ab-initio calculations. Because of unavoidable numerical noise, often equivalent entries have slightly different values, and the final value needs

to be symmetrized somehow. This is performed during the re-symmetrization necessary to address the “sum rules” conservation.

Sum rules and re-symmetrization. Invariance with respect to any global rigid displacement translates into “sum rules” for anharmonic IFCs:

$$\sum_k \Phi(i, j, k)_{\alpha\beta\gamma} = 0, \forall \text{ permutations of } i, j, k. \quad (18)$$

Due to finite size effects, the calculated IFCs are not perfectly symmetric and do not strictly satisfy Eq. (18), causing numerical instabilities. To tackle the issue, different strategies have been proposed such as Lagrange multipliers²⁵ and orthogonalization procedures.⁷⁵ We implement an iterative algorithm which corrects $\Phi(i, j, k)$ and fulfills the constraints.

Given a set of $\Phi(i, j, k)$ the error x of each sum rule at step $\mathcal{N}$ is defined as

$$x(i, j)_{\alpha\beta\gamma}^{\mathcal{N}} \equiv \sum_k \Phi(i, j, k)_{\alpha\beta\gamma}^{\mathcal{N}} \quad (19)$$

Each iteration is composed of correction and re-symmetrization of equivalent IFCs. Correction, $\Phi(i, j, k)_{\alpha\beta\gamma}^{\mathcal{N}} \rightarrow \Phi(i, j, k)_{\alpha\beta\gamma}^{\mathcal{N}+1}$ is given by:

$$\begin{aligned} \Phi(i, j, k)_{\alpha\beta\gamma}^{\mathcal{N}+1} = & (1 - \mu) \Phi(i, j, k)_{\alpha\beta\gamma}^{\mathcal{N}} + \frac{\mu}{n_{\text{eq}}} \\ & \times \sum_{\substack{i'j'k' \\ \alpha'\beta'\gamma'}}^{\text{eq}} \left(\Phi(i', j', k')_{\alpha'\beta'\gamma'}^{\mathcal{N}} - \frac{x(i', j')_{\alpha'\beta'\gamma'}^{\mathcal{N}} |\Phi(i', j', k')_{\alpha'\beta'\gamma'}^{\mathcal{N}}|}{\sum_{k''} |\Phi(i', j', k'')_{\alpha'\beta'\gamma'}^{\mathcal{N}}|} \right), \end{aligned} \quad (20)$$

where the term $x^{\mathcal{N}} |\Phi^{\mathcal{N}}| / \sum |\Phi^{\mathcal{N}}|$ corrects Φ based on the total error times the absolute contribution of Φ in the “sum rule”. The sum over the combination of indices $\{i', j', k', \alpha', \beta', \gamma'\}$ giving IFCs equivalent to $\Phi(i, j, k)_{\alpha\beta\gamma}$ (there are n_{eq}) is meant to symmetrize the error across all the entries. The mixing fraction in the iterative process, μ , can be adjusted by the user to optimize convergence rate and robustness. Overall, with increasing number of neighbor shells, the user can effectively reduce this systematic error and achieve effective convergence of κ_{ℓ} .

Calculation workflows. The calculations parameters and methods are described in the Methods section.

• Anharmonic scattering time $\tau_{\lambda}^{\text{anh}}$:

$$\begin{aligned} \text{AFLOW-AAPL} & \xrightarrow{\text{finite forces}} \psi \xrightarrow{\text{Eq. (17)} \text{ force constants}} \Phi^{\mathcal{N}} \xrightarrow{\text{Eq. (20)} \text{ symmetrization}} \Phi \rightarrow \\ & \xrightarrow{\text{Eq. (14)} \text{ scatt. matrix}} V^{\pm} \xrightarrow{\text{Eqns. (12-13)} \text{ scatt. rates}} \Gamma^{\pm} \xrightarrow{\text{Eq. (11)} \text{ anh. scatt. time}} \tau_{\lambda}^{\text{anh}}. \end{aligned} \quad (21)$$

• Elastic scattering time $\tau_{\lambda}^{\text{iso}}$ (isotopic disorder) and grain boundaries scattering time $\tau_{\lambda}^{\text{bnd}}$ (polycrystalline materials):

$$\begin{aligned} \text{AFLOW-AAPL} & \xrightarrow{\text{ab initio}} \psi \xrightarrow{\text{Eq. (16)} \text{ force const.}} \Phi \xrightarrow{\text{Eq. (4)} \text{ dynamical mat.}} D(\mathbf{q}) \rightarrow \\ & \left\{ \begin{array}{l} \xrightarrow{\text{Eq. (3)} \text{ phonons}} \omega_{\lambda} \xrightarrow{\text{Eq. (8)} \text{ elastic scatt. time, rate}} \tau_{\lambda}^{\text{iso}}, \Gamma_{\lambda\lambda'} \\ \xrightarrow{\text{Eq. (6)} \text{ group velocities}} v_{\lambda} \xrightarrow{\text{Eq. (9)} \text{ grain bound. scatt. time}} \tau_{\lambda}^{\text{bnd}} \end{array} \right. \quad (22) \end{aligned}$$

• Conductivity $\kappa_{\ell}^{\alpha\beta}$:

$$\left\{ \tau_{\lambda}^{\text{anh}}, \tau_{\lambda}^{\text{iso}}, \tau_{\lambda}^{\text{bnd}} \right\} \xrightarrow{\text{Eq. (7)} \text{ total scatt. time}} \tau_{\lambda}^0 \xrightarrow{\text{Eq. (2)} \text{ mean free disp.}} \mathbf{F}_{\lambda} \xrightarrow{\text{Eq. (1)} \text{ conductivity}} \kappa_{\ell}^{\alpha\beta}. \quad (23)$$

Analysis of results. Different statistical parameters are used to measure the qualitative and quantitative agreements of AAPL with respect to experimental values. The Pearson correlation coefficient $r[\{X\}, \{Y\}]$ is a measure of the linear correlation between two variables, $\{X\}$ and $\{Y\}$:

$$r = \frac{\sum_i (X_i - \bar{X})(Y_i - \bar{Y})}{\sqrt{\sum_i (X_i - \bar{X})^2} \sqrt{\sum_i (Y_i - \bar{Y})^2}}, \quad (24)$$

where $\bar{X}$ and $\bar{Y}$ are the averages of $\{X\}$ and $\{Y\}$.

The Spearman rank correlation coefficient $\rho[\{X\}, \{Y\}]$ is a measure of the monotonicity of the relationship between two variables. The values of the two variables $\{X\}$ and $\{Y\}$ are sorted in ascending order, and are assigned rank values $\{x\}$ and $\{y\}$ which are equal to their position in the sorted list. The correlation coefficient is then given by

$$\rho = \frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2} \sqrt{\sum_i (y_i - \bar{y})^2}}. \quad (25)$$

ρ is useful for determining how well the values of one variable can predict the ranking of the other variable.

The root-mean-square relative deviation (RMSrD), of the calculated κ vs. the experiment is also investigated. The RMSrD will measure the quantitative difference between AAPL and experimental results:

$$\text{RMSrD} = \sqrt{\frac{\sum_i \left(\frac{x_i - y_i}{x_i} \right)^2}{N_{\{X, Y\}} - 1}}, \quad (26)$$

Lower values of RMSrD indicate better agreement.

Performance

Scaling. The calculation of the anharmonic IFCs is the most computationally expensive step in the method. First, we test the number of required calculations for some structural prototypes, such as diamond (spacegroup: Fd $\bar{3}$ m, #227; Pearson symbol: cF8; Strukturbericht designation: A4; AFLOW Prototype: A_cF8_227_a²⁶ (http://aflow.org/CrystalDatabase/A_cF8_227_a.html)), rocksalt (Fm $\bar{3}$ m, #225, cF8, B1, AB_cF8_225_a_b²⁶ (http://aflow.org/CrystalDatabase/AB_cF8_225_a_b.html)), fluorite (Fm $\bar{3}$ m, #225, cF12, C1, AB2_cF12_225_a_c²⁶ (http://aflow.org/CrystalDatabase/AB2_cF12_225_a_c.html)), wurtzite, (P6 $_3$ mc, #186, hP4, B4, AB_hP4_186_b_b²⁶ (http://aflow.org/CrystalDatabase/AB_hP4_186_b_b.html)), ZrO $_2$ (P4/nmc, #137, tP6) and corundum (R $\bar{3}$ c, #167, hR10, D5 $_1$, A2B3_hR10_167_c_e²⁶ (http://aflow.org/CrystalDatabase/A2B3_hR10_167_c_e.html)) for which there are abundant available experimental data. The number of calculations and how they scale are compared with respect to the chosen cut-off for the IFCs (see Fig. 1) for different available software (Phono3py and ShengBTE software packages). The number of required static calculations increases with the cell's complexity, the total number of atoms, and the number of inequivalent positions in the primitive cell. AAPL reduces the number of required calculations compared to the other two codes for the six tested prototypes, indicating that the AAPL algorithm is efficient at handling symmetry equivalence. For example, in silicon and using the minimum shell cut-off, AAPL only needs 21 calculations, while ShengBTE requires 76. The advantage is preserved while increasing the range of the interactions. For example, Phono3py requires 616 static calculations for CaF $_2$ with seventh-neighbor shells, whereas AAPL needs less than one-third of this amount (176). Figure 1 summarizes the scaling results.

Fig. 1 Scaling benchmark. Number of required static calculations for **a** Si, **b** NaCl, **c** CaF₂, **d** ZnO, **e** ZrO₂ and **f** Al₂O₃ for the computation of the 3rd order IFCs, applying different cut-offs (n^{th} -neighbor) using AAPL (green), Phono3py (red), and ShengBTE (blue)

Validation with experiments. A data set of 30 compounds is used to validate our framework. The list of materials includes semiconductors and insulators that belong to different structural prototypes, such as diamond (A_cF8_227_a²⁶), rocksalt (AB_cF8_225_a_b²⁶), and fluorite (AB2_cF12_225_a_c²⁶). To maximize the heterogeneity of the data set, materials are selected containing as many different elements as possible from the *s*-blocks, *p*-blocks, and *d*-blocks of the periodic table. The comparison of calculated vs. experimental values of κ_L is summarized in Table 1 and Fig. 2.

Different statistical quantities are used to measure qualitative and quantitative agreements between the AAPL and experimental results (Table 2). AAPL results strongly correlate with experimental findings, with relatively small RMSrD from experiment demonstrating the reliability and robustness of the framework. The algorithm should not be blamed for systematic errors in the ab-initio characterization of the compounds (such as the ones containing Pb).

AAPL is also compared with approximate phenomenological frameworks, such as AFLOW-AGL¹⁵ and AFLOW-QHA-APL^{7,86}. Qualitatively, all the three frameworks have high linear correlation with experiments (Pearson, *r*); AAPL and QHA-APL are also very effective in rank ordering the compounds (Spearman, *p*). Quantitatively, AAPL has the lowest RMSrD value, followed by QHA-APL and AGL. Accuracy strongly correlates with computational costs (AAPL $\gg$ QHA-APL $>$ AGL), so that the users can choose which technique best fulfills their screening needs.

Single-crystal and nanocrystalline silicon. Silicon is the perfect benchmark for testing the reliability of AAPL: extensive availability

of experimental data for well-characterized samples^{87,78} and limited computational cost due to the diamond crystal structure with two atoms in the primitive cell and fcc lattice. Figure 3a depicts the calculated lattice thermal conductivity at different temperatures for single-crystal and polycrystalline samples compared to single-crystal experimental values from ref. 78. The calculations published by Carbogno et al. using the Green-Kubo formalism are also included⁸⁸. Boundary effects can be included in two ways: i. by calculating the complete $\kappa_L^{ab}(L)$ (workflow (23)) for average grain sizes having different $\tau_\lambda^{\text{bnd}}(L)$ (workflow (22)) or ii. by neglecting $\tau_\lambda^{\text{bnd}}$ from the total scattering time τ_λ^0 (Eq. (7)) and restricting the summation to the modes with a mean free path shorter than L ($\Lambda < L$, Eq. (10)): $\kappa_{L,(\Lambda < L)}^{ab}$. Both approaches are implemented in AAPL. Comparison with experimental values for different polycrystalline Si samples (average grain size $L = 64, 76, 80, 155, 550$ and $20,000$ nm) at 300 K are presented in Fig. 3b. Both approximations of grain boundary scattering effects show the same trend and are very close to the experimental results, corroborating the validity of our approaches.

Non-cubic systems. The lattice thermal conductivity is addressed for three well-characterized systems.

- i. Tetragonal zirconia (*t*-ZrO₂) is used for energy and biomedical applications because of its combination of strength, fracture toughness, and ionic conductivity, as well as a low thermal conductivity coating material for protecting metals from excessive heat. Some properties strictly depend on the stabilization of the tetragonal phase as the monoclinic polymorph (*m*-ZrO₂) is the stable phase at room

Table 1. Calculated and experimental lattice thermal conductivity of diamond (Strukturbericht: A4; AFLOW standardized prototype name A_cF8_227_a²⁶), rocksalt (B1, AB_cF8_225_a_b²⁶), and fluorite (C1, AB2_cF12_225_a_c²⁶) structure semiconductors and insulators at 300 K

Formula	Pearson	S.G.#	Struk. ^a	$\kappa_{\lambda}^{\text{AAPL}}$	$\kappa_{\lambda}^{\text{Exp}}$
C	cF8	227	A4	2270	2200 ^{76,77}
Si	cF8	227	A4	144	146 ⁷⁸
Ge	cF8	227	A4	46.17	58 ⁷⁹
AgCl	cF8	225	B1	0.67	1 ⁸⁰
BaO	cF8	225	B1	2.99	2.3 ⁸⁰
CaO	cF8	225	B1	20.41	27 ⁸⁰
KBr	cF8	225	B1	2.38	3.4 ⁸⁰
KCl	cF8	225	B1	3.77	7.1 ⁸⁰
KI	cF8	225	B1	1.46	2.6 ⁸⁰
LiF	cF8	225	B1	10.73	17.6 ⁸⁰
LiH	cF8	225	B1	28.4	15 ⁸⁰
MgO	cF8	225	B1	54.06	60 ⁸⁰
NaBr	cF8	225	B1	2.74	2.8 ⁸⁰
NaCl	cF8	225	B1	6.53	7.1 ⁸⁰
NaF	cF8	225	B1	21.11	16.5 ⁸⁰
Nal	cF8	225	B1	1.46	1.8 ⁸⁰
PbS	cF8	225	B1	1.35	2.9 ⁸⁰
PbSe	cF8	225	B1	1.21	2.0 ⁸⁰
PbTe	cF8	225	B1	1.73	2.5 ⁸⁰
RbBr	cF8	225	B1	1.68	3.8 ⁸⁰
RbI	cF8	225	B1	1.64	2.3 ⁸⁰
SrO	cF8	225	B1	9.12	12 ⁸⁰
CdF2	cF12	225	C1	3.01	4.30 ⁸¹
SrCl2	cF12	225	C1	1.80	2.3 ⁸²
Mg2Si	cF12	225	C1	15.67	18.8 ⁸³
Mg2Ge	cF12	225	C1	11.49	15.7 ⁸³
Mg2Sn	cF12	225	C1	9.91	11.1 ⁸³
Mg2Pb	cF12	225	C1	9.20	18 ⁸³
CaF2	cF12	225	C1	7.04	9.76 ⁸¹
CeO2	cF12	225	C1	11.35	10.8 ⁸⁴
ThO2	cF12	225	C1	14.42	14 ⁸⁵

Units: κ_{ℓ} in W/(m K)
^a Strukturbericht
^b κ_{ℓ} at 200 K

temperature.⁹⁸ Stabilization is performed by alloying the system with aliovalent ions, such as yttrium.⁹⁸ Carbogno et al. recently calculated κ_{ℓ} for $t\text{-ZrO}_2$ with a combination of ab-initio molecular dynamics and Green–Kubo formalism.⁸⁸ Figure 3c shows a comparison: AAPL values (PBE and LDA functionals) for $t\text{-ZrO}_2$ and $m\text{-ZrO}_2$, Carbogno et al. for $t\text{-ZrO}_2$, and the experimental values for $m\text{-ZrO}_2$ and yttrium-stabilized $t\text{-ZrO}_2$ (YSZ).^{89,90} Methods using molecular dynamics with ions responding “classically” to ab-initio forces tend to overestimate the thermal conductivity below the Debye temperature. Classical dynamics follows the Dulong–Petit limit for the specific heat—Boltzmann distribution of phonon energies—and the quantum of transferred heat is unavoidably overestimated (κ_{ℓ} increases with C_v). On the contrary, force-constants methods have phonons energies following the Bose–Einstein distribution by construction (Eq. (1)). They

Fig. 2 Comparison with experiments of different AFLOW techniques. Calculated lattice thermal conductivities at 300 K vs. experimental. Blue circles are used for AAPL results, empty orange triangles for the quick AFLOW—AGL prediction of refs. 15,29, and empty green squares for AFLOW—QHA-APL results of ref. 7. The red line represents equality (calculation = experiments)

Table 2. Root mean square relative deviation (RMSrD), and Pearson and Spearman correlations for the material data set

	AGL	QHA-APL	AAPL
r	0.997	0.999	0.999
ρ	0.706	0.976	0.933
RMSrD	86.20%	58.16%	27.57%

might neglect higher-order phonons’ renormalization effects, thus producing underestimated κ_{ℓ} .⁹⁹ Above the Debye temperature (~ 590 K for ZrO_2 ^{100,101}), the phonons mean free path reduces and the statistical distributions become classic. In that regime, the two methods will approach the correct experimental values from different directions.

- Wurtzite zinc oxide (ZnO) is another well-characterized system. Oluranyolemi et al.⁹¹ and Barrado et al.⁹² reported κ_{ℓ} of polycrystalline ZnO with an average particle size of 20 and 1000 nm, respectively. AAPL, performed with average grain size of 500 nm, is in very good agreement with the experimental results capturing the effects of the grain boundaries’ scattering (Fig. 3d).
- Rhombohedral aluminum oxide, $\alpha\text{-Al}_2\text{O}_3$, which contains 30 atoms in its conventional unit cell, is chosen to test the framework’s robustness. To the best of our knowledge, this is the largest unit cell ever characterized for lattice thermal conductivity with ab-initio means (FeSb3, containing 16 atoms was studied in ref. 102). AAPL results are again in excellent agreement with experimental values⁹³ for a wide range of temperatures (Fig. 3d).

Extension to ACBN0 pseudo-hybrid functional. The accuracy of the results ultimately relies on the quality of the computed IFCs with ab-initio. The use of hybrid functionals¹⁰³ or advanced electronic structure methods, such as GW¹⁰⁴ to compute the IFCs is limited^{105,106} because of their computational costs. It was also reported that hybrid functionals can predict false lattice instability in some cases.¹⁰⁷ Recently, the ACBN0 functional was introduced in order to facilitate the accurate characterization of electronic properties of correlated materials.⁴⁰ ACBN0 is a pseudo-hybrid Hubbard density functional that introduces a new self-consistent ab-initio approach to compute U without the need for empirical

Fig. 3 Comparison with experiments for selected materials systems. **a** Calculated lattice thermal conductivity for single-crystal (blue) and nanocrystalline silicon with different grain sizes. Blue circles represent measurements for single-crystal Si from ref. 78. **b** Cumulative lattice thermal conductivity, $\kappa_{L,(\Lambda \leq L)}$, (green) of Si as a function of the average grain size, L , at 300 K. Lattice thermal conductivity (orange) including the scattering of phonons due to grain boundaries (see Eq. (9)) is also presented. Blue circles represent experimental data from ref. 87. **c** Lattice thermal conductivity of t -ZrO₂ (blue lines) and m -ZrO₂ (green lines) using AAPL and Green-Kubo formalism⁸⁸ (orange). Blue circles and green squares represent experimental data from refs. 89, 90. **d** Calculated lattice thermal conductivity for ZnO (green) and α -Al₂O₃. Blue circles and green squares represent experimental data from refs. 91–93. **e** Lattice thermal conductivity of CaF₂ within the ACBN0 method (green) and PBE functional (orange). Blue circles represent experimental data from ref. 94. **f** Phonon dispersion of CaF₂ within the ACBN0 method (green). The PBE phonon dispersion (orange) is also shown for comparison. Blue triangles and open squares represent neutron scattering data from refs. 95,96, respectively. Purple diamonds represent Raman and infrared data from ref. 97

parameters. ACBN0 can improve not only the description of the electronic structure, but also the prediction of the structural and the vibrational parameters of solids.⁴⁵ One of the reasons for this is the better prediction of the Born charges, $\mathbf{Z}$, and the dielectric tensor, ϵ_{∞} , compared to LDA or GGA functionals.⁴⁵ If the ACBN0 functional improves the vibrational parameters of solids, it can be assumed that the calculations of other temperature-dependent properties, such as κ_L , may be improved too. As a testbed, calcium fluoride, CaF₂, is chosen, because of the ample available experimental data.^{81,95–97} CaF₂ has been extensively used in

optical devices due to its low refractive index, wide band gap, low dispersion, and large broadband radiation transmittance.^{108–110}

A package implementing a minimalist AFLOW-Python approach to high-throughput ab-initio calculations for the generation of tight-binding hamiltonians and the calculation with the ACBN0 functional (AFLOW π)¹¹¹ is used to obtain the ACBN0 electronic structure of CaF₂. A U_{eff} of 13.43 is obtained for F- p orbitals. This value is then used inside AAPL for the rest of the calculations. AAPL + ACBN0 almost perfectly predicts the experimental κ_L in contrast with AAPL + PBE which greatly underestimates κ_L over

the entire temperature range (Fig. 3e). Phonon band structures have also been calculated with the harmonic library (APL) to explain the difference (Fig. 3f). ACBN0 reproduces the phonon dispersion better than the PBE functional. PBE, as a GGA functional, overestimates bond length and hence it tends to underestimate vibrational frequencies.¹¹² On the contrary, ACBN0 describes the bond length more accurately, obtaining frequencies higher than PBE and closer to the experimental values.⁹⁵ Major differences between ACBN0 and PBE results come from the optical bands, so the two main properties that are involved in the splitting of the optical band due to the non-analytical contributions to the dynamical matrix are compared. While the Born charges are similar for ACBN0 and PBE (2.33 e and 2.34 e, respectively), there are significant differences in the high-frequency dielectric constant ϵ_∞ (2.083 and 2.305, respectively). The value obtained using ACBN0 is closer to the experimental ϵ_∞ (2.045)⁹⁷ than that obtained using PBE.

CONCLUSIONS

In this article, we have presented the AAPL, which was developed to compute the third-order IFCs and solve the BTE within the high-throughput AFLOW framework. The software automatically predicts the lattice thermal conductivity of single-crystals and polycrystalline materials by using a single input file and no further user intervention. The symmetry analysis has been optimized to further reduce the number of static calculations compared to other packages. The robustness and accuracy of the code have been tested with a set of 30 materials that belong to different space groups. APL has been combined with the ACBN0 pseudo-hybrid functional to predict the lattice thermal conductivity of CaF_2 . Our results demonstrate that using ACBN0 can improve not only the electronic structure description of the material compared to the GGA functional, but also phonon-dependent properties, such as the thermal conductivity.

METHODS

Geometry optimization

All structures are fully relaxed using the automated framework AFLOW^{28–39} and the VASP package.¹¹³ Optimizations are performed following the AFLOW standards.³⁴ The PAW potentials¹¹⁴ are used and the exchange and correlation functionals parameterized by the generalized gradient approximation proposed by Perdew–Burke–Ernzerhof (PBE).¹¹⁵ All calculations use a high-energy cut-off, which is 40% larger than the maximum recommended cut-off among all component potentials, and a **k**-point mesh of 8000 **k**-points per reciprocal atom. Primitive cells are fully relaxed (lattice parameters and ionic positions) until the energy difference between two consecutive ionic steps is smaller than 10^{-4} eV and forces in each atom are below 10^{-3} eV/Å.

Phonon calculations

Phonon calculations are performed using the Automatic Phonon Library, APL, as implemented in AFLOW, and by using VASP to obtain the second-order IFCs via the finite-displacement approach.⁸⁶ The magnitude of the displacement is 0.015 Å. Electronic self-consistent field iterations for static calculations are stopped when the difference of energy between the last two steps is less than 10^{-5} meV. The threshold ensures a good convergence for the wavefunction and sufficiently accurate values for forces and harmonic constants. Non-analytic contributions to the dynamical matrix are also included using the formulation developed by Wang et al.⁵⁷ Frequencies and other related phonon properties are calculated on a $21 \times 21 \times 21$ **q**-point mesh in the Brillouin zone, which is a tradeoff between the computational cost, convergence of the phonon density of states, pDOS, and the derived thermodynamic properties. Integrations within the Brillouin zone are obtained by using the linear interpolation tetrahedron method available in AFLOW.

Lattice thermal conductivity

Anharmonic force constants are extracted from a $4 \times 4 \times 4$ supercell using a cut-off that includes all 4th-neighbor shells. Thermal conductivity is evaluated on a $21 \times 21 \times 21$ **q**-point mesh using $\zeta = 0.1$ for the Gaussian smoothing, Eq. (15). The dense mesh ensures the convergence of the values obtained for κ_i .²⁵ The IFC calculations are iterated self-consistently until all sum rules are lesser than 10^{-7} eV/Å³.

Acronyms in the framework

The following acronyms are used in the article. AAPL: Automatic-Anharmonic-Phonon-Library; AGL: AFLOW-Gibbs-Library;¹⁵ APL: Automatic-Phonon-Library;^{28,35,16} QHA: quasi-harmonic approximation;⁷ ACBN0: Agapito Curtarolo Buongiorno Nardelli ab-initio DFT functional;⁴⁰ AFLOW π : A minimalist AFLOW-Python approach to high-throughput ab-initio calculations for the generation of tight-binding hamiltonians and the calculation with the ACBN0 functional.¹¹¹

Data availability

All the data and codes are freely available to the public as part of the AFLOW online repository and can be accessed through www.aflow.org.

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AUTHOR CONTRIBUTIONS

J.J.P. developed and implemented the AAPL framework within AFLOW, with advice and assistance from J.C., C.T., P.N., M.d.J., M.A., and S.C. D.U., P.N. and J.J.P. ran the ab-initio calculations to obtain the lattice thermal conductivity. M.F. and M.B.N. performed the ACBN0 calculations to obtain the self-consistent *U* values for CaF_2 . All authors contributed to the analysis of the results and the writing of the paper.

ADDITIONAL INFORMATION

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