Automating Anharmonic Lattice Thermal Conductivity Calculations using Compressed Sensing Lattice Dynamics

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Abstract

Engineering heat transfer is critical for applications in heat exchangers, semiconductor devices, thermoelectrics and more. This demand motivates a high-throughput computational methodology for systematically designing materials with improved thermal properties. One emerging method which can rapidly and explicitly consider phonon-phonon interactions even for highly anharmonic materials is Compressive Sensing Lattice Dynamics (CSLD). In fact, CSLD accurately predicts 2nd (harmonic) and 3rd+ order (anharmonic) interatomic force constants (IFCs) with orders of magnitude fewer Density Functional Theory (DFT) calculations than conventional methods. However, doing CSLD can be an exhaustive, many-step process requiring an intimate knowledge of its sensitivity to parameters. Consequently, this work implements an automatic CSLD workflow capable of obtaining the thermal conductivity of potentially thousands of materials, benchmarks the stability of the workflow against materials with a range of anharmonicity, and begins constructing a dataset of thermal conductivity values and phonon dispersion curves to be stored in the Materials Project for public use.

Compressed Sensing Lattice Dynamics

- Phonon-phonon interactions described by anharmonic interatomic force constants (IFC)
- CSLD solves L1 regularized empirical risk minimization to choose sparse set of IFCs that reproduce atomic forces

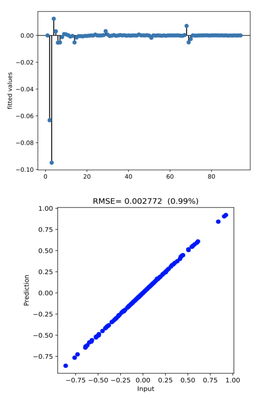
$$V = V_0 + \phi_a^i u_a^i + \frac{\phi_{ab}^{ij}}{2} u_a^i u_b^j + \frac{\phi_{abc}^{ijk}}{3!} u_a^i u_b^j u_c^k + \dots$$

$$F_a^i = -\frac{\partial V}{\partial u_a^i} = -\phi_a^i - \frac{\phi_{ab}^{ij}}{2} u_b^j - \frac{\phi_{abc}^{ijk}}{3!} u_b^j u_c^k - \dots$$

$$\mathrm{A} = \begin{bmatrix} -1 & -\frac{1}{2}u_{b,1} & -\frac{1}{3!}u_{b,1}u_{c,1} \\ \dots & \dots & \dots \\ -1 & -\frac{1}{2}u_{b,L} & -\frac{1}{3!}u_{b,L}u_{c,L} \end{bmatrix}$$

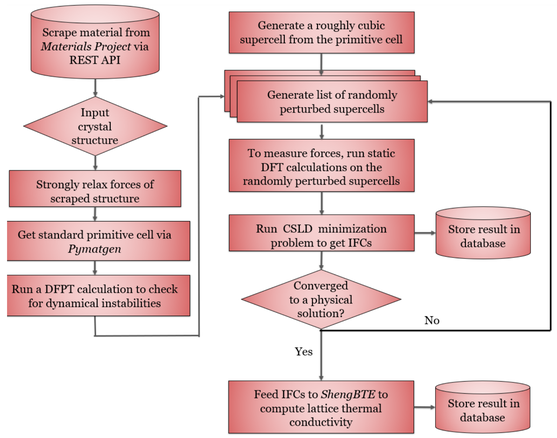
$$\phi^{CS} = \underset{\phi}{\operatorname{argmin}} ||\phi||_1 + \mu||\mathbb{F} - A\phi||_2^2$$

Shown right (top) is a plot of fitted IFC values versus IFC indices and (bottom) a comparison of forces predicted by CSLD-generated IFCs versus a holdout set of forces from a DFT calculation on cubic silicon.



Workflow

Outline for the automated workflow implemented in atomate for calculating interatomic force constants, phonon dispersion curves, and lattice thermal conductivity considering up to 3rd order anharmonicity. If the CSLD step does not converge to a physical solution, then perturbed supercells with larger atomic displacements will be dynamically generated to account for the possibility of the structure having loosely bound atoms. If CSLD then still fails to converge, an error is raised.

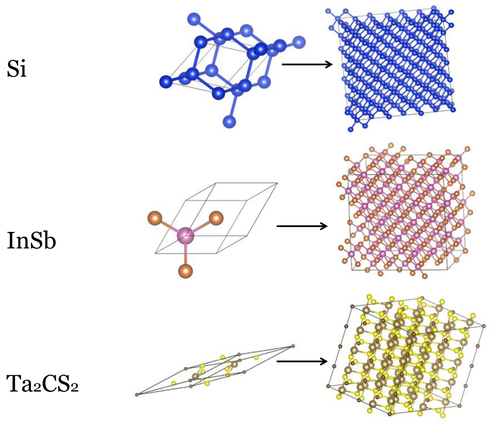


Automatically generating input for CSLD

Three new features were implemented into *Pymatgen*.

CubicSupercellTransformation

- Input a crystal structure, a number of nearest neighbor distances that the supercell must encapsulate, and bounds on the number of atoms in the supercell
- Iteratively solves for valid supercell lattice vectors that satisfy all constraints
- Outputs a transformation matrix and a supercell structure to encapsulate physically relevant interatomic clusters



PerturbSitesTransformation

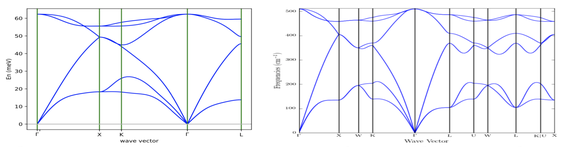
- Input a list of displacement distances and a crystal structure
- Output a list of supercells with randomly perturbed atomic sites to generate atomic force data for CSLD fitting

ShengBTE IO

- A Python adaptor for the *ShengBTE*, a software that can calculate lattice thermal conductivity from IFCs

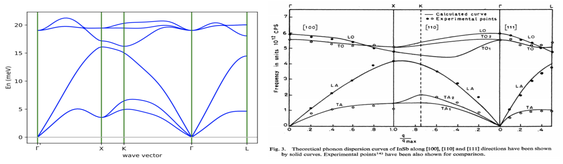
Benchmarking the workflow

Si



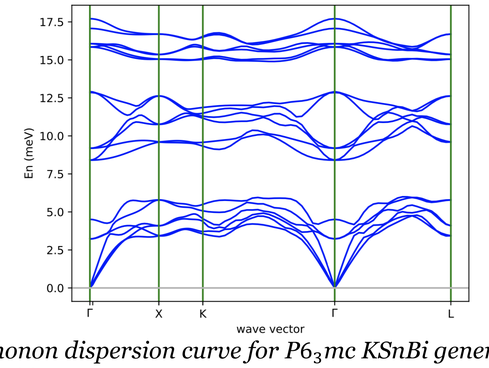
Phonon dispersion curve for Fd-3m silicon generated by CSLD (left) and by Abinit (right).

InSb



Phonon dispersion curve for F-43m indium antimonide generated by CSLD (left) and by neutron scattering experiments (right) [1].

KSnBi



Phonon dispersion curve for $P6_3mc$ KSnBi generated by CSLD.

[1] Ram, R. & Kushwaha, S. Phonon Dispersion Relations for GaAs and InSb. *J. Phys. Soc. Japan* **54**, 617–624 (1985)

Next Steps

- Run the workflow on a large number of materials from the Materials Project database
- Validate CSLD-generated dispersion curves with Density Functional Perturbation Theory
- Use sequential optimizers to search for interesting materials (i.e. thermal barrier coatings, thermoelectrics, etc.)
- Generate data for the Materials Project for public reference
- Data mine generated data to uncover hidden empiricisms and clustering
- Interpretable machine learning to discover important factors for predicting lattice thermal conductivity

Try the workflow!

CSLD:
https://github.com/LLNL/csld

atomate:
https://github.com/hackingmaterials/atomate

Pymatgen:
https://github.com/materialsproject/pymatgen

Fireworks:
https://github.com/materialsproject/fireworks

Custodian:
https://github.com/materialsproject/custodian

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