Unlocking Phonon Properties of a Large and Diverse Set of Cubic Crystals by Indirect Bottom-up Machine Learning Approach

Alejandro Rodriguez,[1] Changpeng Lin,[2] Chen Shen,[3] Kunpeng Yuan,[4] Mohammed Al-Fahdi,[1] Xiaoliang Zhang,[4] Hongbin Zhang,[3] and Ming Hu[1,*]

[1]*Department of Mechanical Engineering, University of South Carolina, Columbia, SC 29208, USA*

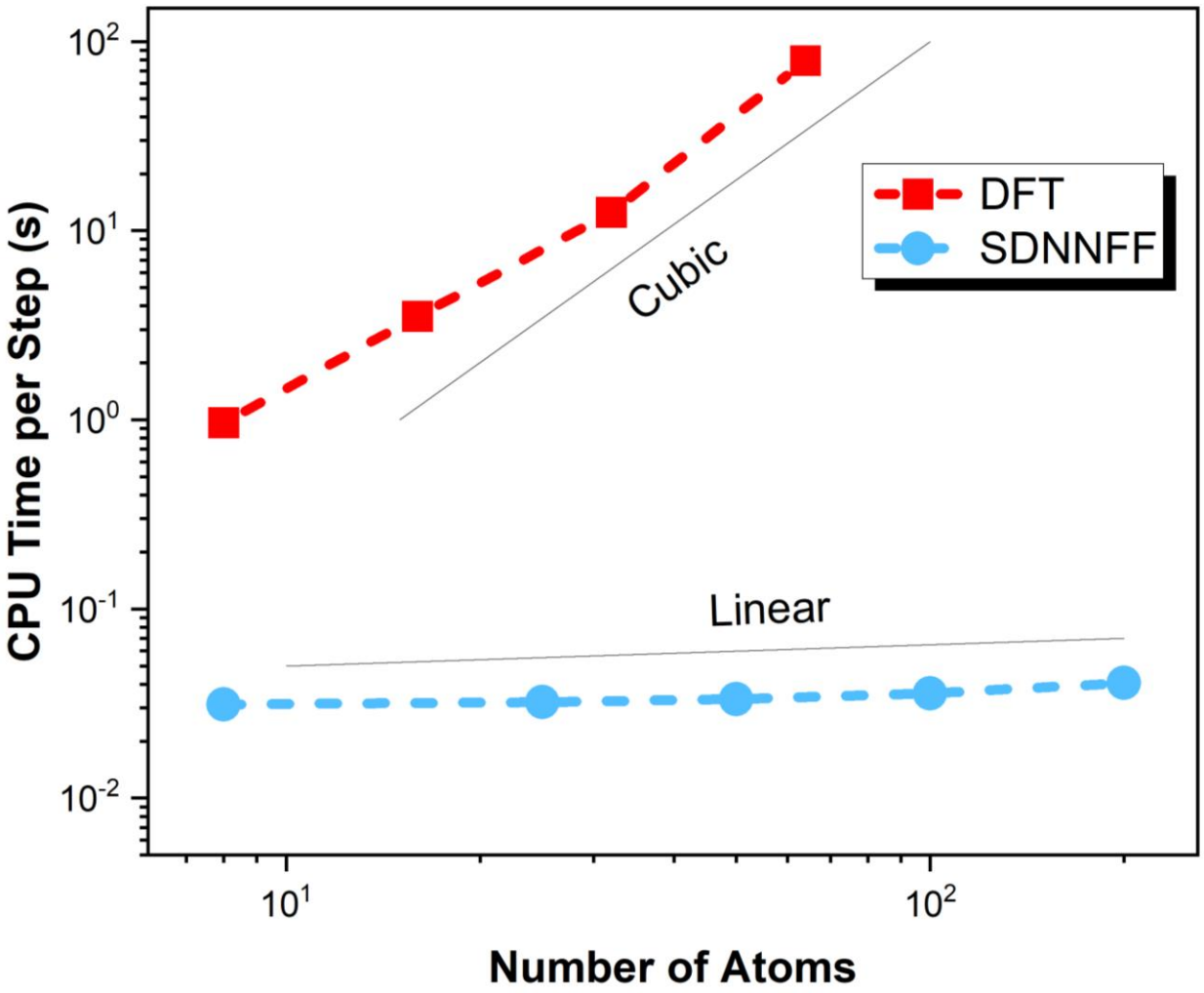
[2]*Theory and Simulation of Materials (THEOS), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland*

[3]*Institute of Materials Science, Technical University of Darmstadt, Darmstadt 64287, Germany*

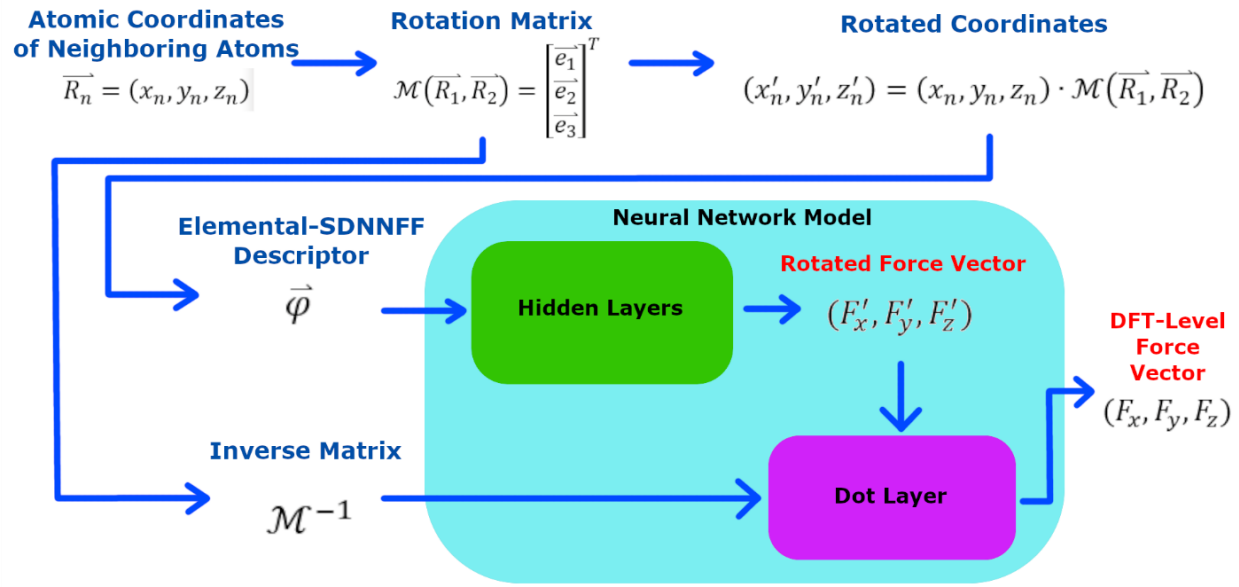
[4]*School of Energy and Power Engineering, Dalian University of Technology, Dalian 116024, China*

[*] Author to whom all correspondence should be addressed. E-Mail: hu@sc.edu (M.H.)

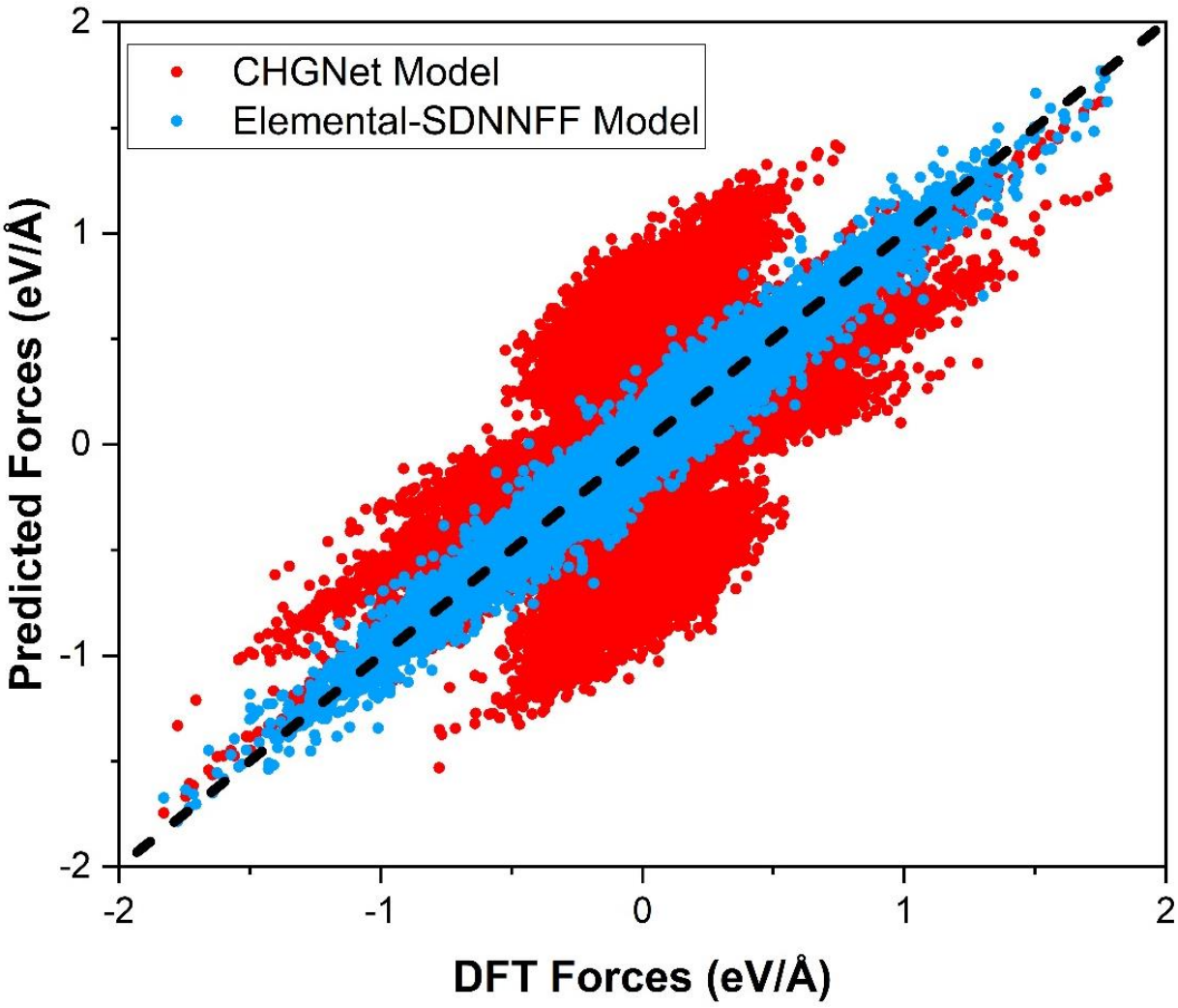
Supplementary Figures



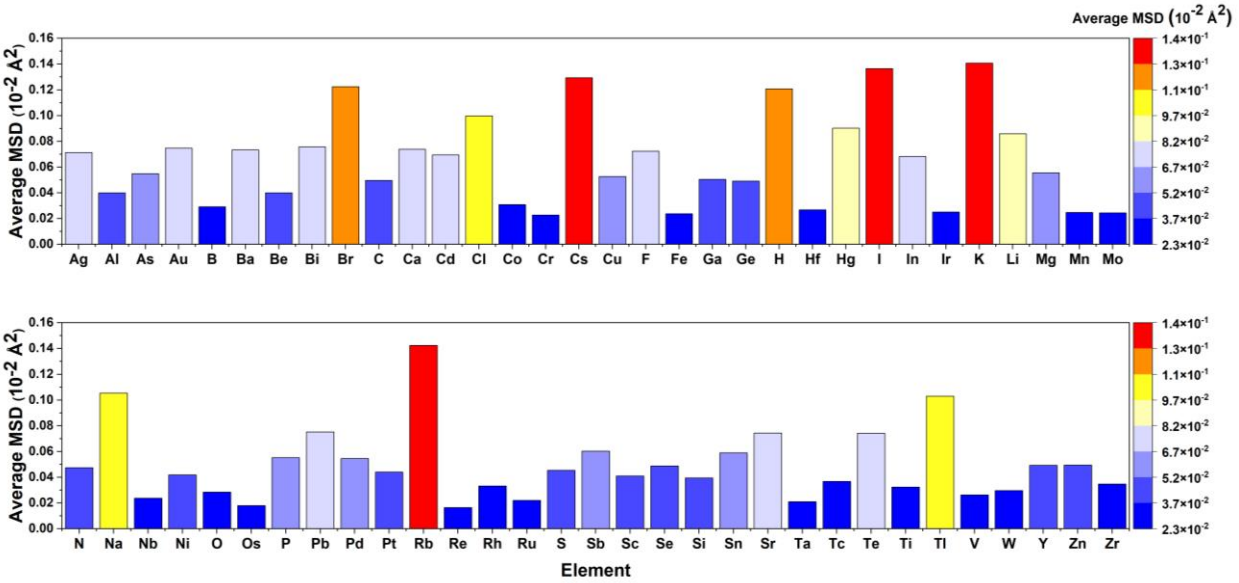
Supplementary Figure S1. The CPU time per step against the number of atoms in the system from DFT and the SDNNFF model. Lines labeled 'linear' and 'cubic' are shown for guidance.



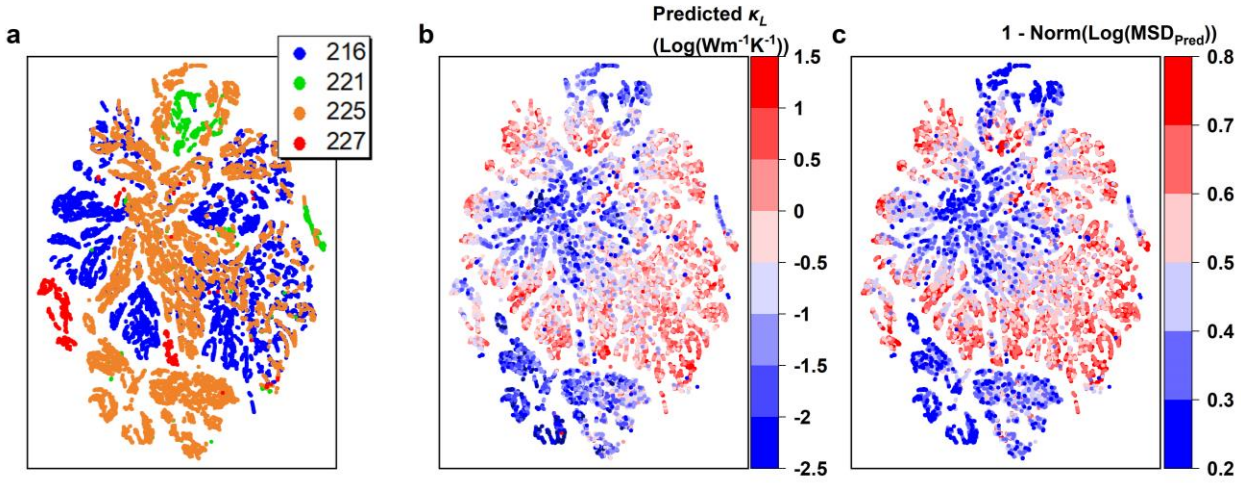
Supplementary Figure S2. Schematic of our elemental neural network architecture considering rotational covariance. The basic input for the neural network model is the representation of local atomic environment with given cutoff distance, while the output is the atomic forces along three cartesian coordinates directions.



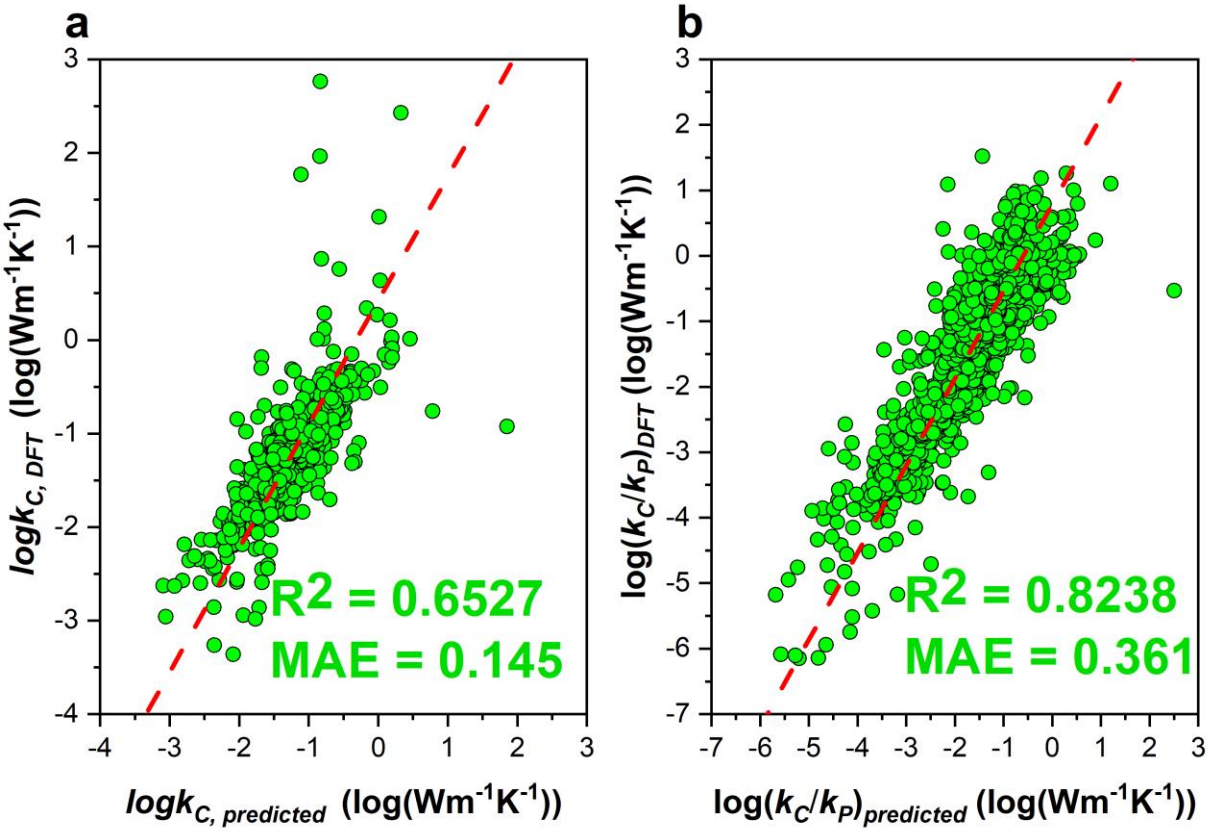
Supplementary Figure S3. Comparison of the atomic forces in 400 untrained structures between prediction models, CHGNet[1] and Elemental-SDNNFF, and DFT. The structures span half, full, quaternary Heusler structures, and single and double perovskites. The RMSE for the Elemental-SDNNFF and CHGNet models are 29.3 meV/Å and 121 meV/Å, respectively. The black dashed line represents perfect agreement.



Supplementary Figure S4. Average mean square displacement (MSD) across 25,901 predicted stable structures. The color bar represents the magnitude of the average MSD in Å^2.



Supplementary Figure S5. t-SNE plots of Elemental-SDNNFF structural input vectors of 25,901 structures colored according to (a) space group number, (b) predicted κ_L at 300K, and (c) normalized log(max MSD) at 300 K subtracted from unity.



Supplementary Figure S6. (left) The off-diagonal contribution to lattice thermal conductivity as compared between the DFT and predicted results by Elemental-SDNNFF model computed for 2,398 structures from the training pool. (right) The ratio of off-diagonal contribution to the lattice thermal conductivity without considering off-diagonal term for the same structures.

Supplementary References

1. Deng, B. *et al.* CHGNet: Pretrained universal neural network potential for charge-informed atomistic modeling. 1–12 (2023).