

# Unleashing the Power of Artificial Intelligence for Phonon Thermal Transport

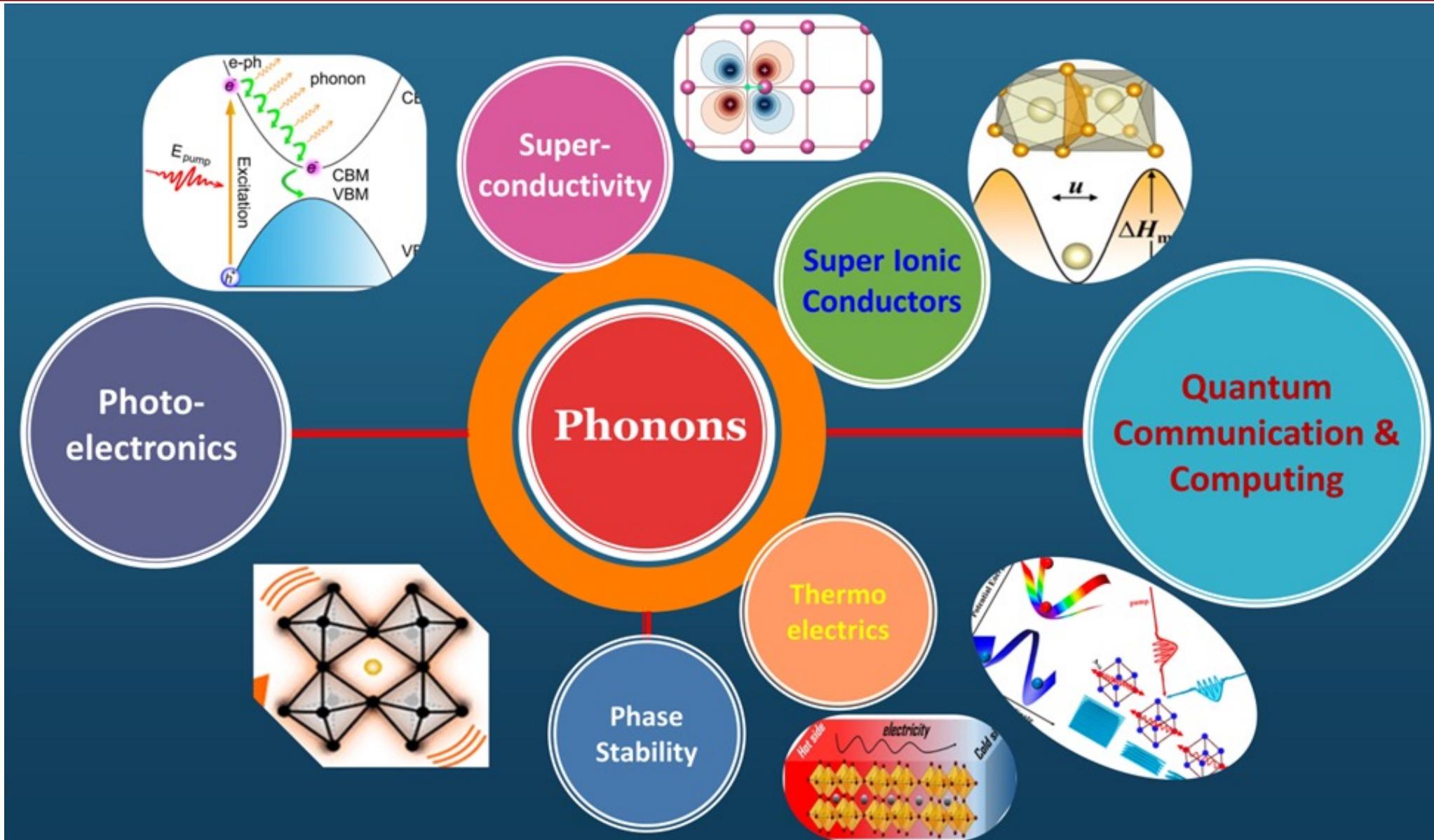
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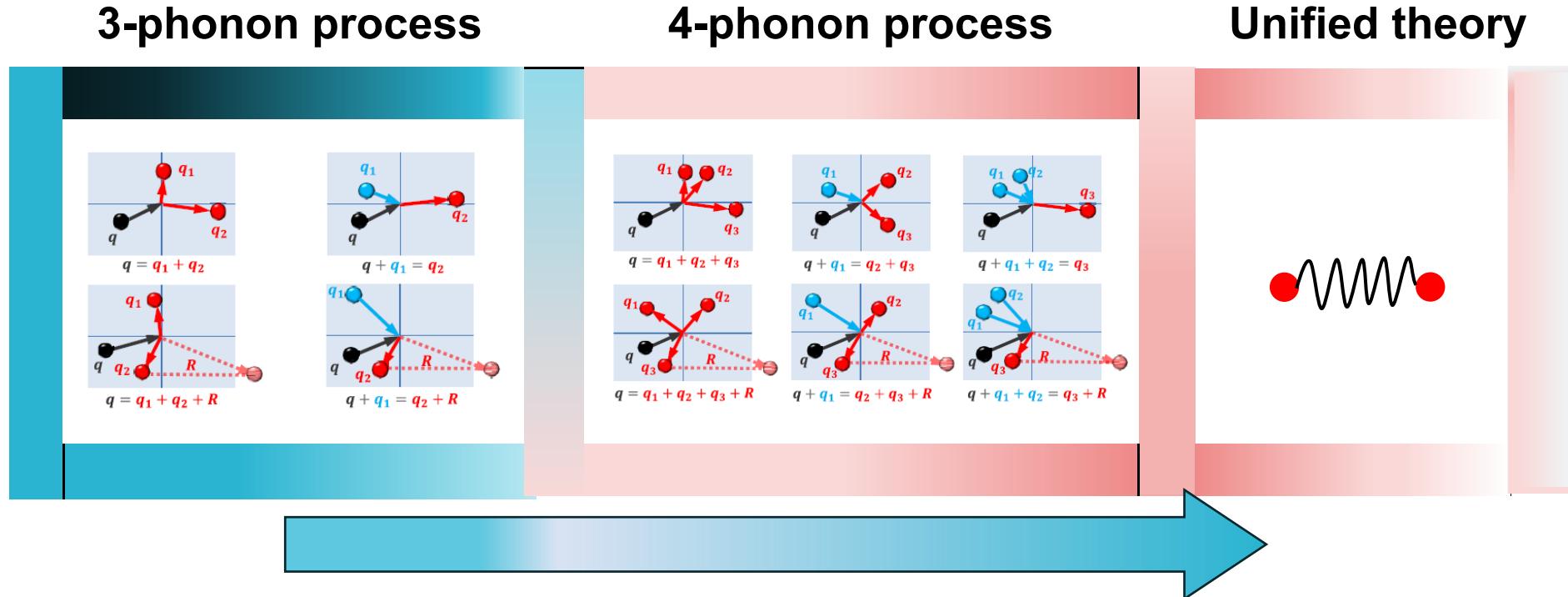


# OUTLINE

- ❑ Current Status of Data-Driven Approach for Thermal Science
  - Direct ML models with and without predefined descriptors
- ❑ Our Approach for Large-scale Comprehensive Phonon Transport Prediction
  - Our indirect bottom-up Elemental-SDNNFF algorithm
  - Pushing the phonon thermal transport to the limit
- ❑ Summary & Outlook



# ➤ Recent development of phonon transport theory



## References:

*Computer Physics Communications*, **185**, 1747 (2014)

*Physical Review B*, **97**, 045202 (2018)

*Computer Physics Communications*, **270**, 108179 (2022)

*Nature Physics*, **15**, 809 (2019); *Physical Review X*, **12**, 041011 (2022)



# ➤ State-of-the-art DFT Calculations of Phonons

- Phonon Boltzmann Transport Equation (BTE):

$$\frac{df_\lambda}{dt} = \left. \frac{\partial f_\lambda}{\partial t} \right|_{\text{diffusion}} + \left. \frac{\partial f_\lambda}{\partial t} \right|_{\text{scattering}} = 0$$

- Lattice Dynamics:

- $\Phi_{ij}^{\alpha\beta} \rightarrow 2^{\text{nd}} \text{ order IFCs}$
- $\Psi_{ijk}^{\alpha\beta\gamma} \rightarrow 3^{\text{rd}} \text{ Order IFCs}$
- **Both computed from atomic forces**

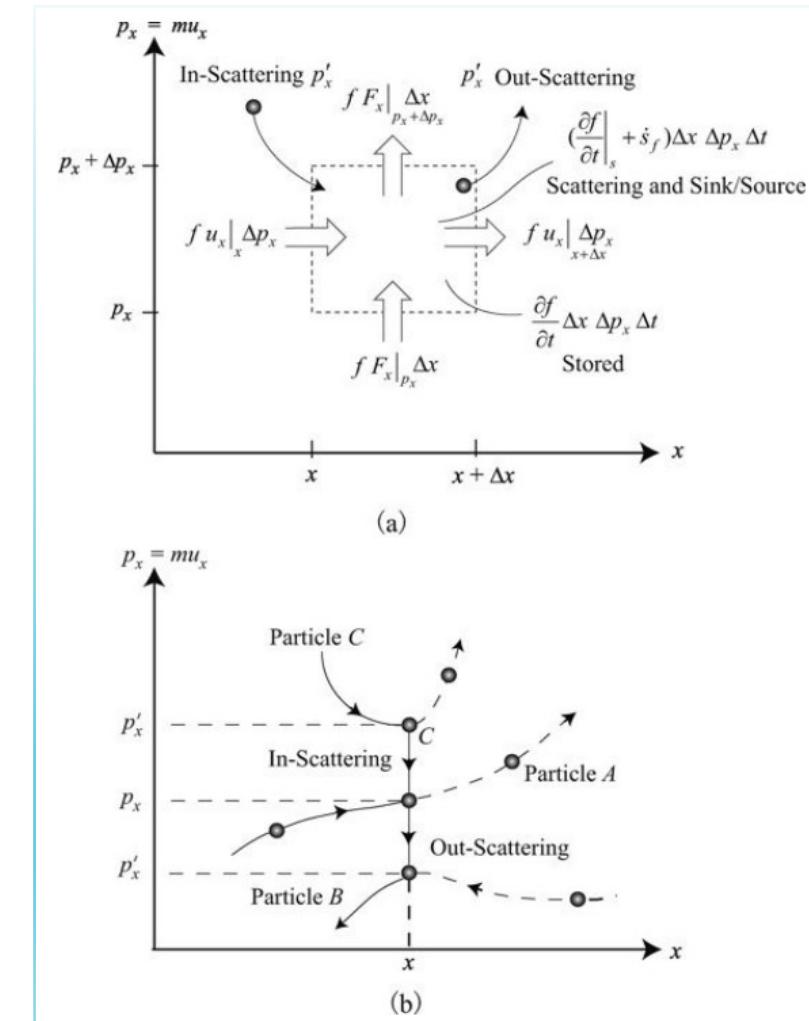
$$V = \sum_{ij,\alpha\beta} \left. \frac{\partial^2 V}{\partial u_\alpha^i \partial u_\beta^j} u_\alpha^i u_\beta^j \right|_{eq} + \sum_{ijk,\alpha\beta\gamma} \left. \frac{\partial^3 V}{\partial u_\alpha^i \partial u_\beta^j \partial u_\gamma^k} u_\alpha^i u_\beta^j u_\gamma^k \right|_{eq} + \sum_{ijkl,\alpha\beta\gamma\sigma} \left. \frac{\partial^4 V}{\partial u_\alpha^i \partial u_\beta^j \partial u_\gamma^k \partial u_\sigma^l} u_\alpha^i u_\beta^j u_\gamma^k u_\sigma^l \right|_{eq} + \dots$$

$\omega$   
noninteracting harmonic

$\omega$   
 $\omega'$   
third-order anharmonic

$\omega$   
 $\omega'$   
 $\omega''$   
 $\omega'''$   
fourth-order anharmonic

$$\kappa_{ph,\alpha} = \sum_{\mathbf{q},v} c(\mathbf{q}) v_{g,\alpha}^2(\mathbf{q}) \tau_\alpha(\mathbf{q})$$



# ➤Challenge: Construct Force Constants by DFT

$$\kappa_{ph,\alpha} = \sum_{\mathbf{q},v} c(\mathbf{q}) v_{g,\alpha}^2(\mathbf{q}) \tau_\alpha(\mathbf{q})$$



3<sup>rd</sup> order IFCs require  
 $\propto N_1 \times N_2 \times N_3 \times n^2$  DFT calculations Needed!

$$\Phi_{ijk}^{\alpha\beta\gamma} = \frac{\partial^3 E}{\partial r_i^\alpha \partial r_j^\beta \partial r_k^\gamma}$$

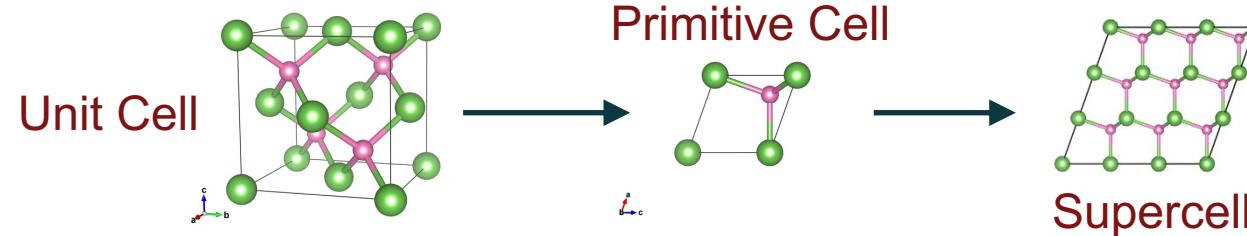
$$\simeq \frac{1}{2h} \left[ \frac{\partial^2 E}{\partial r_j^\beta \partial r_k^\gamma}(r_i^\alpha = h) - \frac{\partial^2 E}{\partial r_j^\beta \partial r_k^\gamma}(r_i^\alpha = -h) \right]$$

$$\simeq \frac{1}{4h^2} \left[ -F_k^\gamma(r_i^\alpha = h, r_j^\beta = h) + F_k^\gamma(r_i^\alpha = h, r_j^\beta = -h) \right.$$

$$\left. + F_k^\gamma(r_i^\alpha = -h, r_j^\beta = h) - F_k^\gamma(r_i^\alpha = -h, r_j^\beta = -h) \right]$$

The supercell-based, finite-difference method (FDM) to calculate the third-order IFCs

*The lower symmetry and the larger number of atoms in the system, the more cases needed for calculating the third-order IFCs*

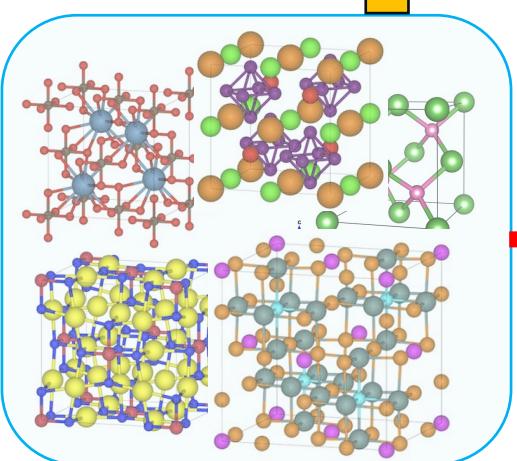


362 DFT: ~17,000 core hours  
for 3x3x3 supercells

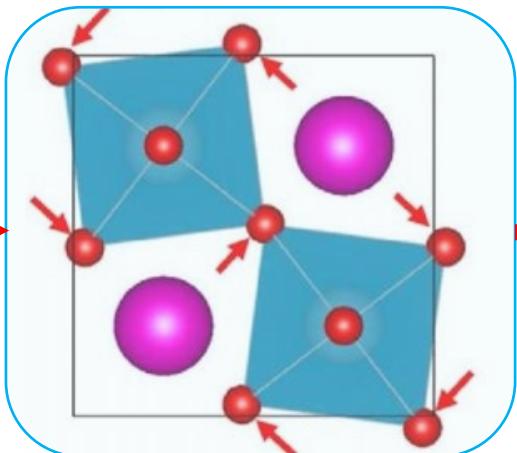


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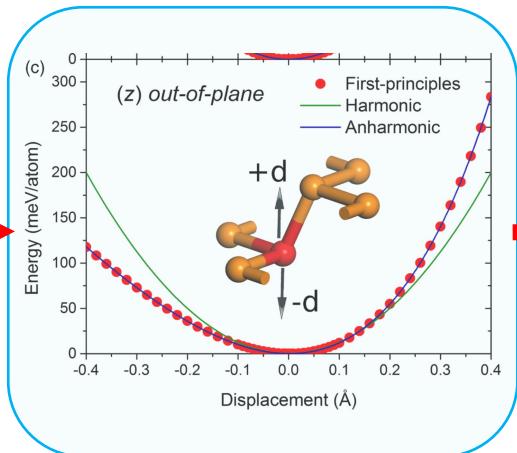
## Direct Method *AI/ML Training & Forward Prediction*



Initial Large Pool of Materials



Supercells with Million-Scale Atomic Environments



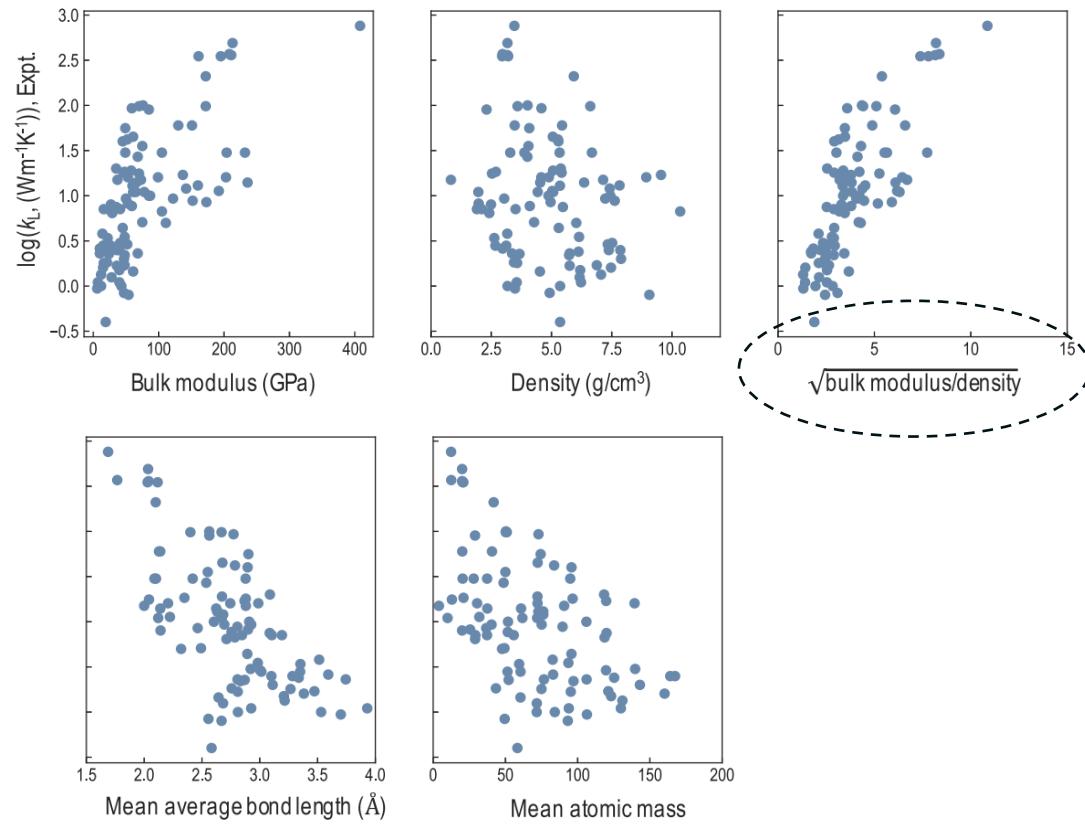
Interatomic Force Constants Fitting

- Phonon Dispersions
- Thermal Conductivity
- Relaxation Time
- Heat Capacity
- Scattering Channels
- Two-Channel Transport
- Phonon Hydrodynamics
- Mechanical Stability
- .....

Comprehensive Phonon Properties

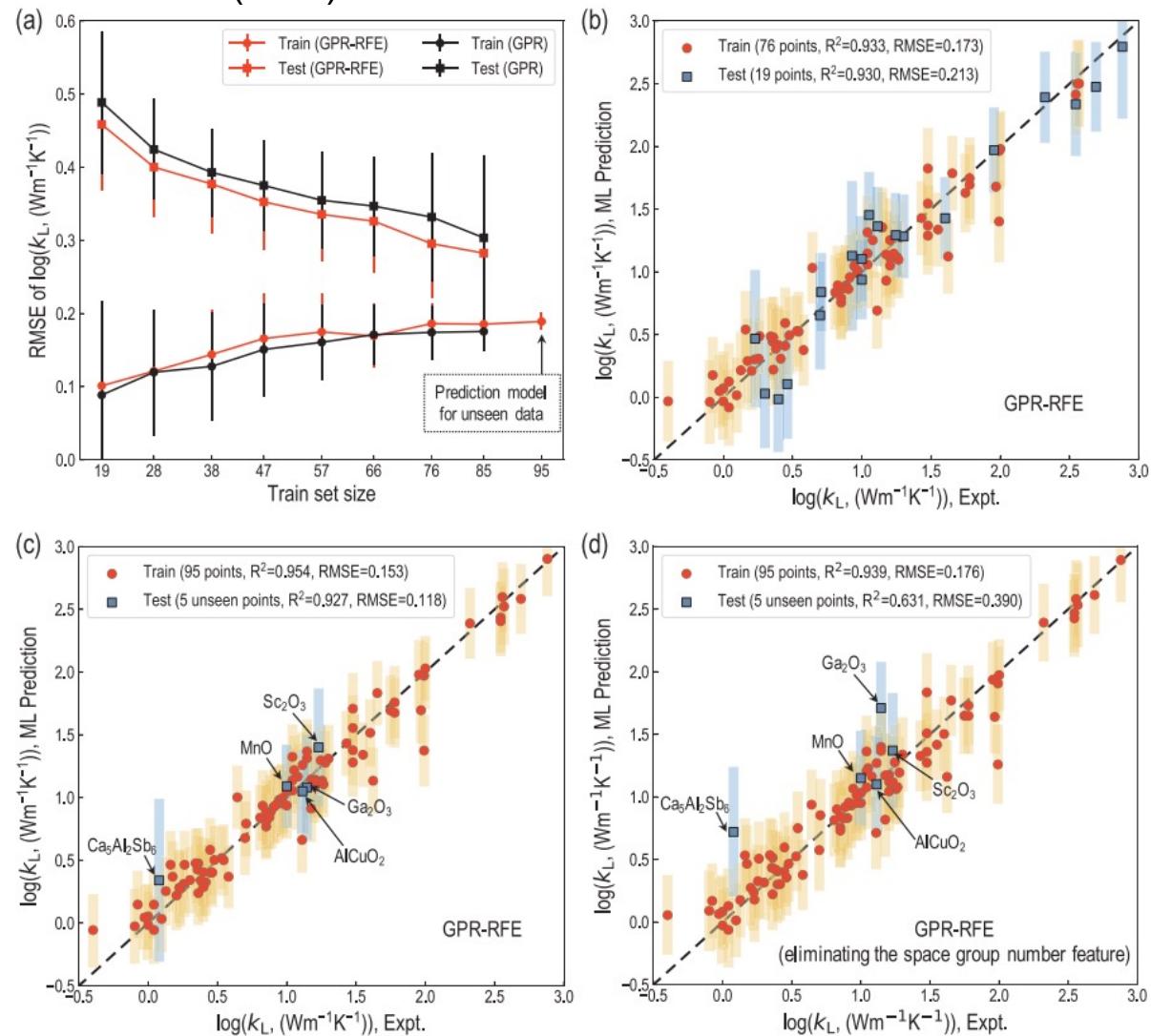


# A benchmark data set of experimental LTC of ~100 inorganic materials

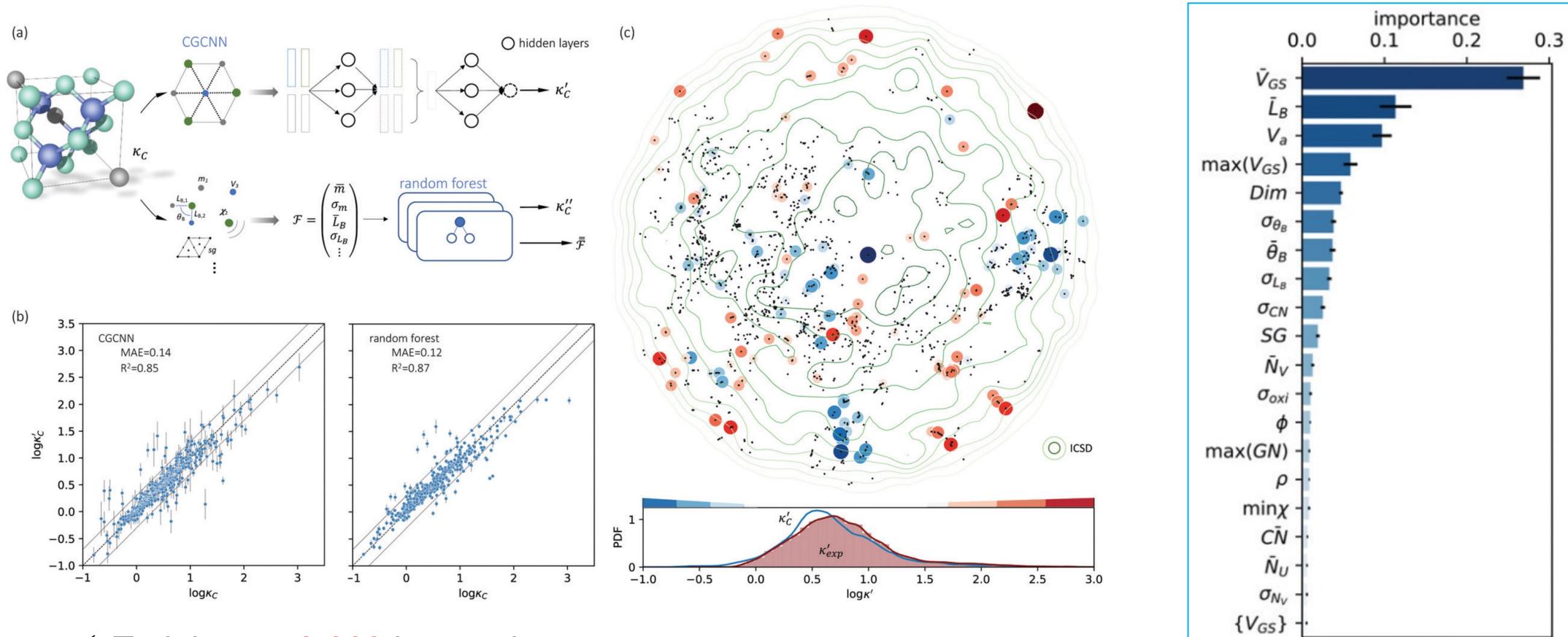


Correlation between experimental LTC and five representative features

## Gaussian process regression (GPR) with radial basis function (RBF) kernel



# ➤ CGCNN Model for Training & Predicting LTC



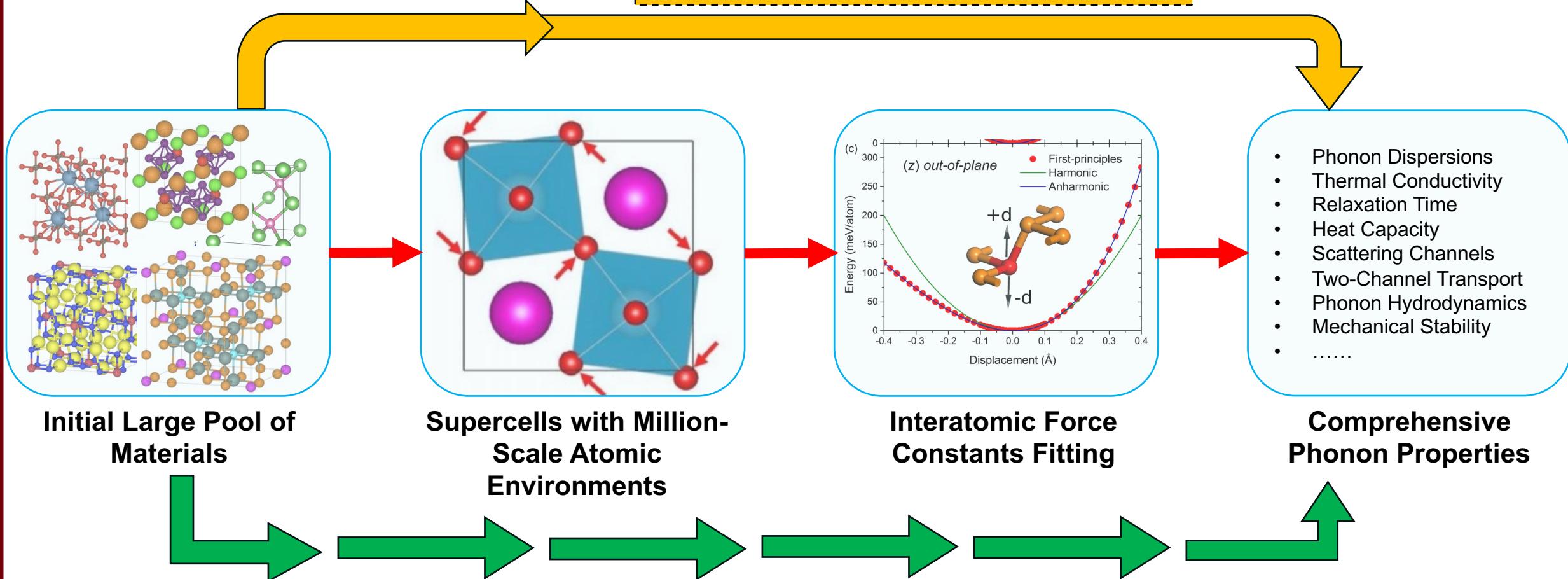
- ✓ Training on 2,668 inorganic structures
- ✓ Predicting LTC of 92,919 entries in ICSD
- ✓ Crystal graph convolutional neural network + random forest models



**Big issue: limited data & lack of comprehensive properties**

## Direct Method

*AI/ML Training & Forward Prediction*



## Indirect Method

Where is big data?

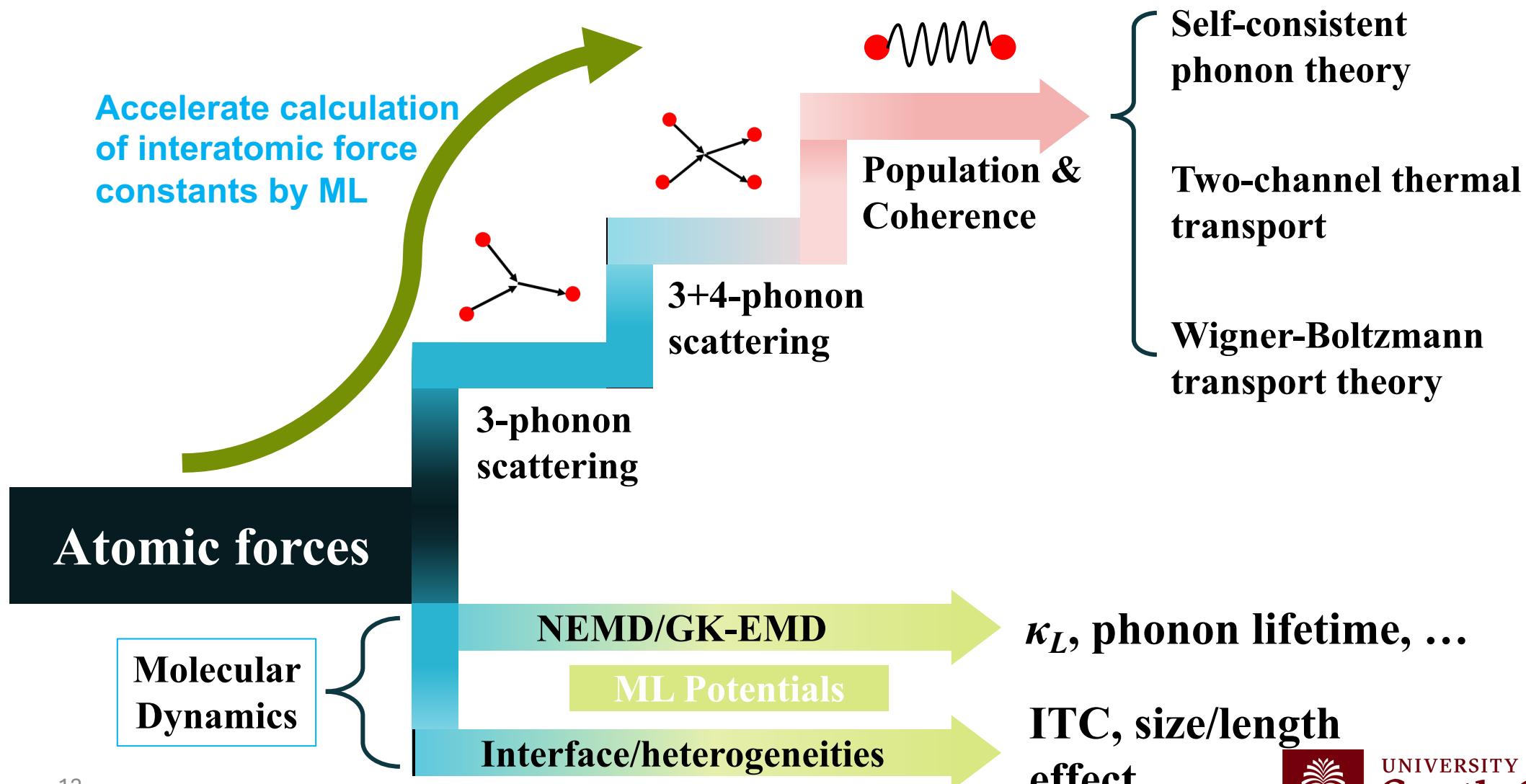


# OUTLINE

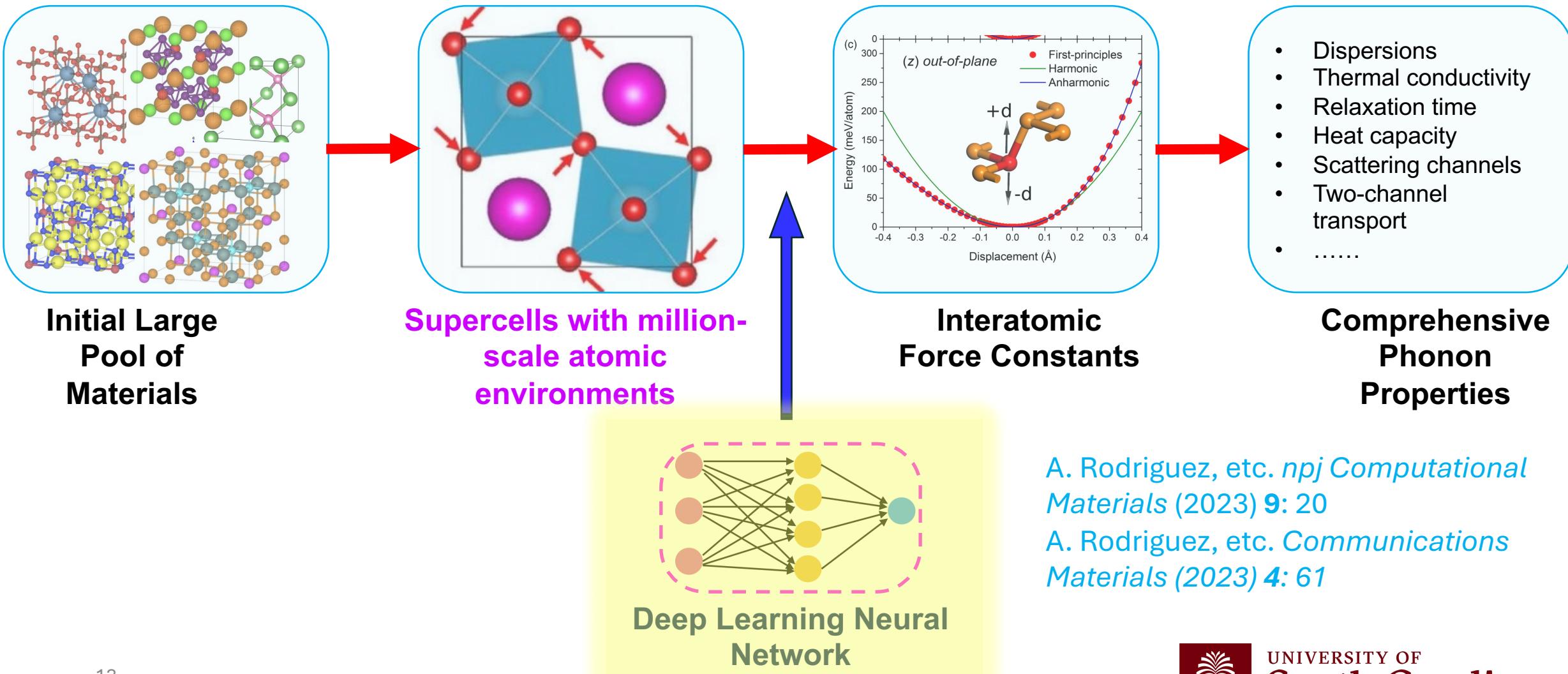
- Current Status of Data-Driven Approach for Thermal Science
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# ➤ Critical Role of Accurately Predicting Atomic Forces for Phonons

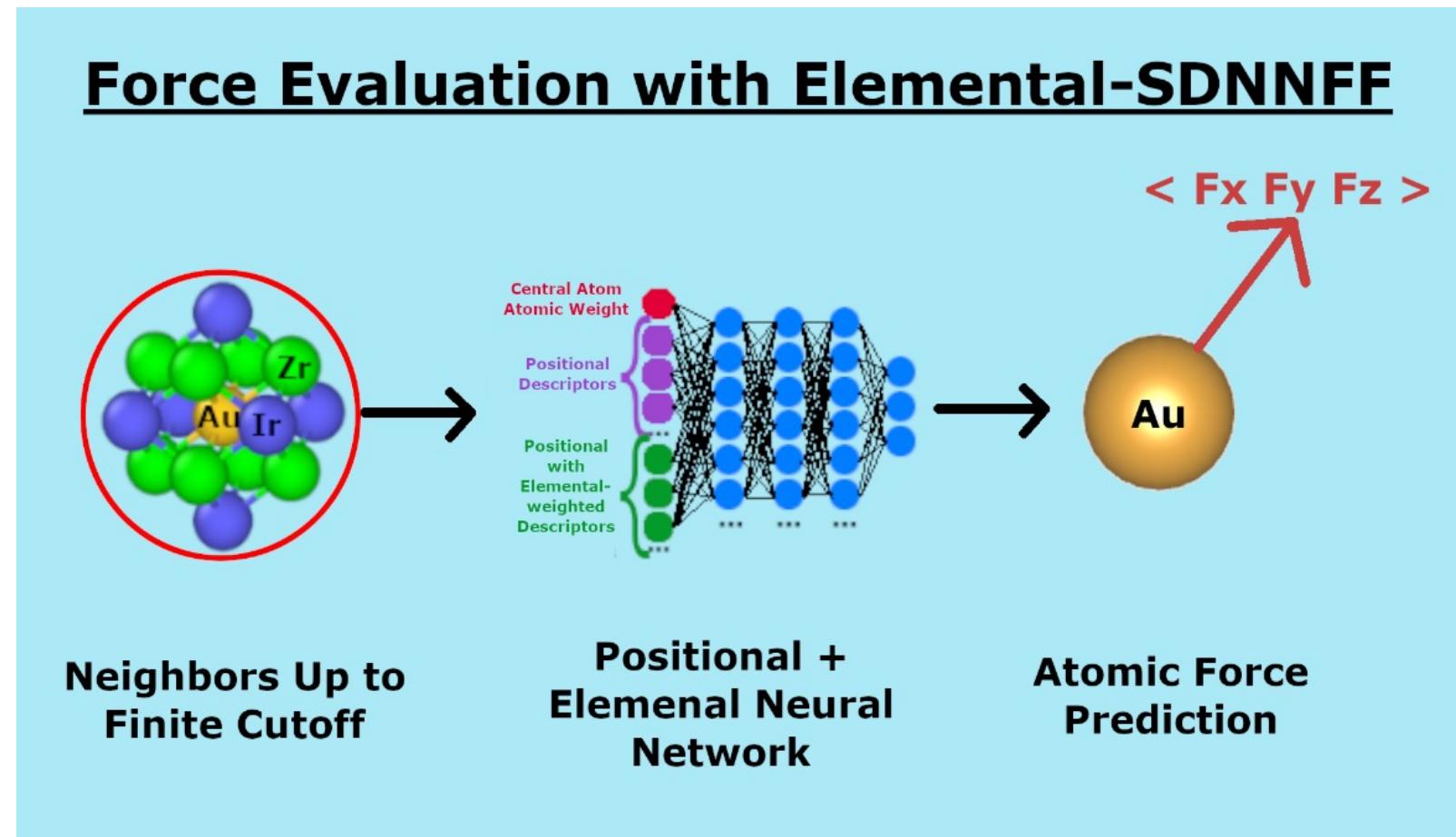


# ➤ An Indirect Bottom-up Machine Learning Approach for Phonons



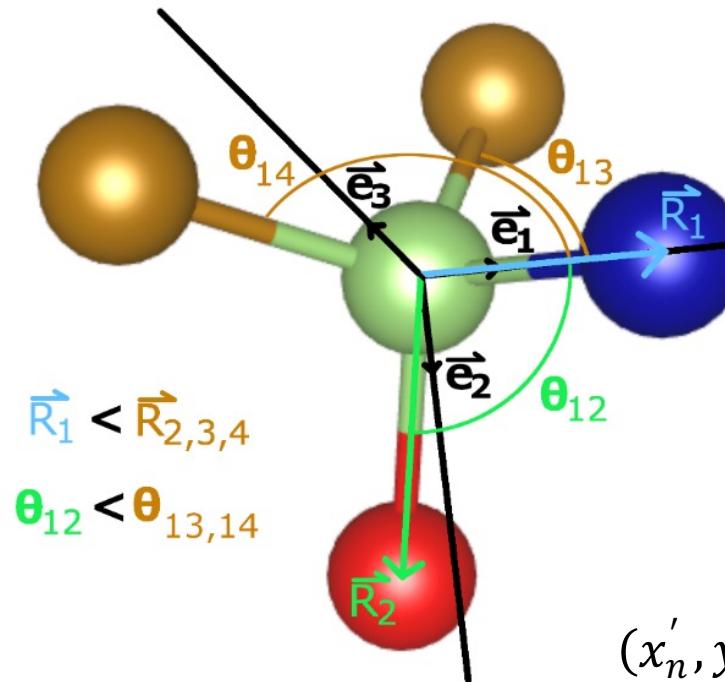
# ➤ Elemental-SDNNFF: Add Elements to SDNNFF

- How to consider species and avoid  $N_{elem}^2$  scaling?
  - *Add second set of positional descriptors weighted by elements*
    - The weights are atomic masses of the element
  - To avoid  $N_{elem}$  networks, dedicate one input to central atom atomic mass
  - Result: *A single NNFF capable of resolving spatial and elemental descriptions of atomic environments*
  - Cost: 2x+1 input descriptors in comparison to original SDNNFF
- 
- *The improved Elemental-SDNNFF can handle elements spanning the periodic table*



# ROTATIONAL COVARIANCE

- Problem:** Rotation of system changes  
3D-dependent descriptor → **Network**  
**cannot use previous knowledge**
- Add rotational covariance
  - Descriptor before rotation equals that after rotation
  - Forces differ by simple rotation matrix  $\mathcal{M}$
- Develop  $\mathcal{M}$  from two neighboring atoms
- The first atom is the closest atom
- The second atom follows the rule:
  - must be coordinated
  - must own smallest angle w.r.t. central and first atom
- Reduce variability of data → **transferability of knowledge**



$$\vec{e}_1 = e(\vec{R}_1)$$

$$\vec{e}_2 = e(\vec{R}_2 - (\vec{R}_2 \cdot \vec{e}_1)\vec{e}_1)$$

$$\vec{e}_3 = \vec{e}_1 \times \vec{e}_2$$

$$\mathcal{M}(\vec{R}_1, \vec{R}_2) = \begin{bmatrix} \vec{e}_1 \\ \vec{e}_2 \\ \vec{e}_3 \end{bmatrix}^T$$

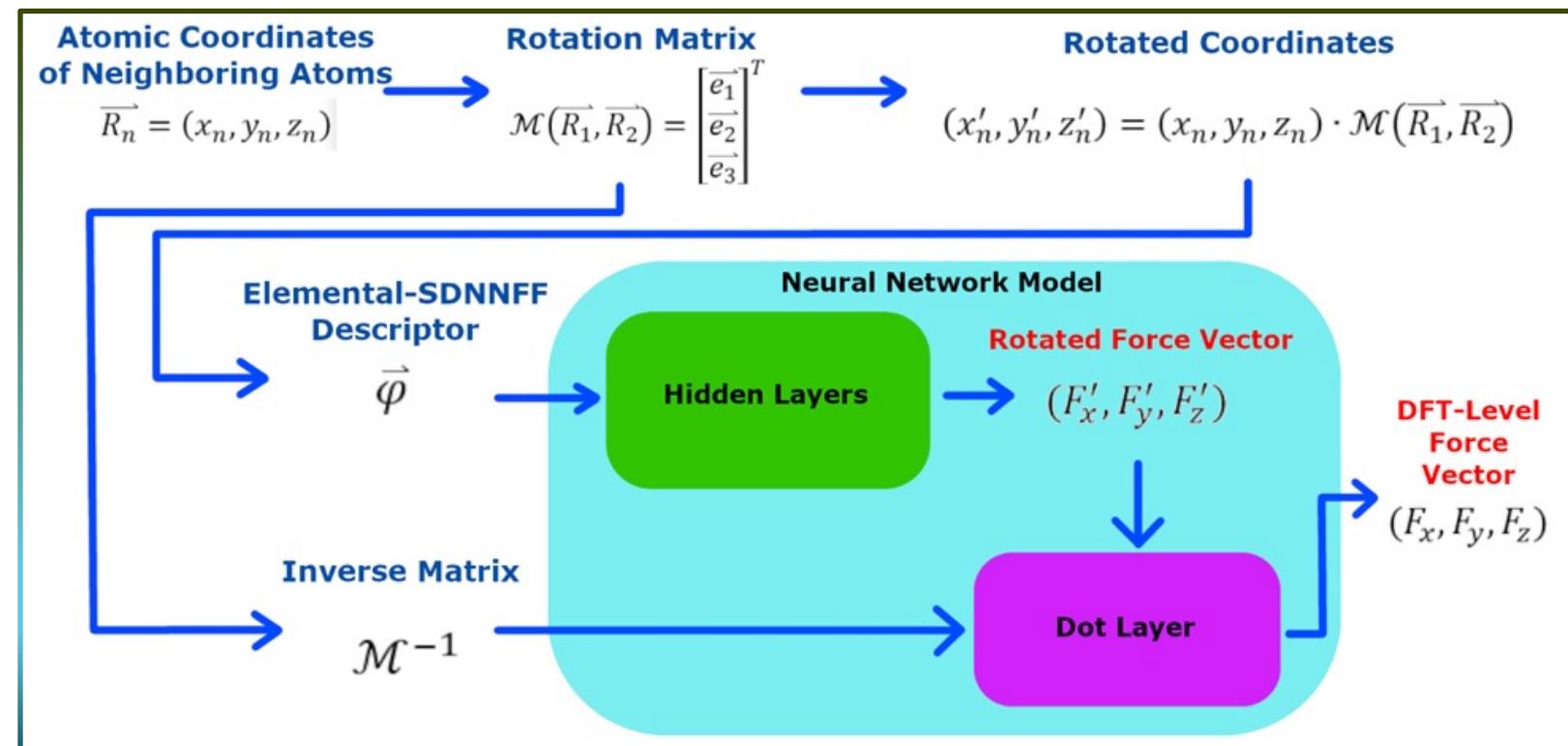
$$(x_n^{'}, y_n^{'}, z_n^{'}) = (x_n, y_n, z_n) \cdot \mathcal{M}(\vec{R}_1, \vec{R}_2)$$



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# ➤ Elemental-SDNNFF: Rotational Covariance

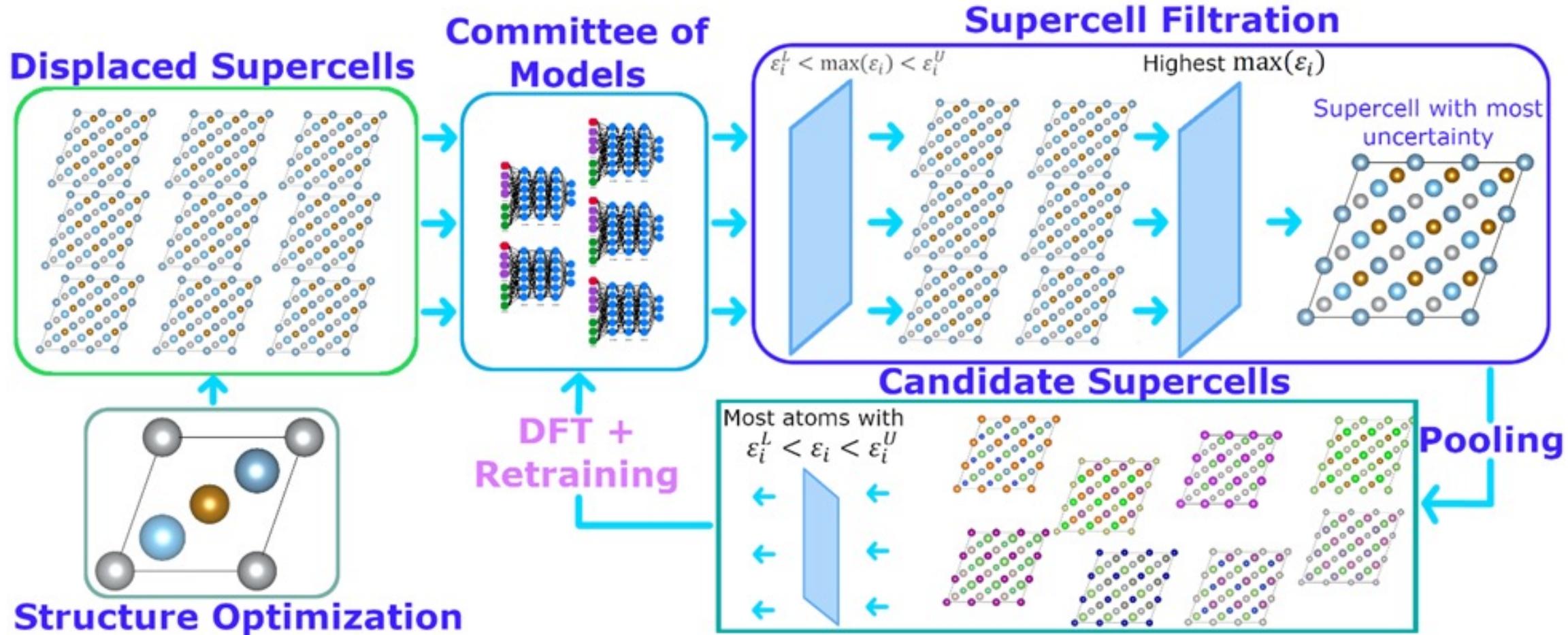
- The advantage of rotational covariance is the capacity to model infinitely many possible rotations with fewer equivalent representations, reducing the redundancy in training similar but rotated atomic systems.
- Rotational covariance also helps reduce the number of DFT configurations needed for force accuracy convergence since redundant atomic neighborhoods of existing but rotated systems are already considered.



- We use the data augmentation to aid the model to understand changes in local environment based on the implemented rotational covariance.*

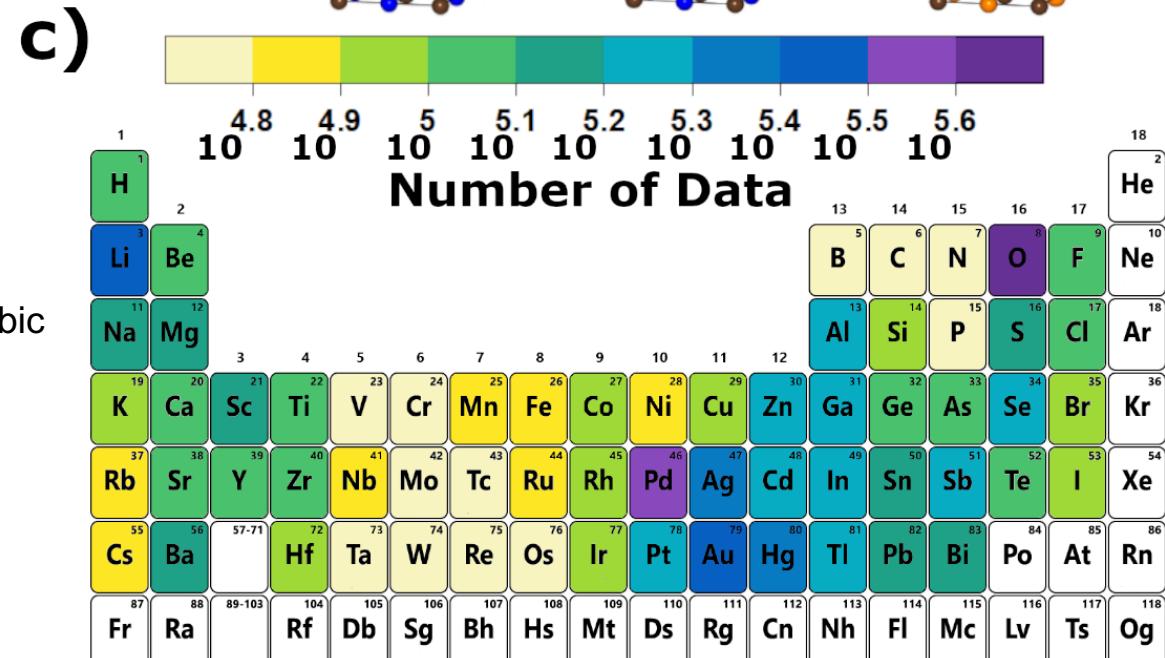
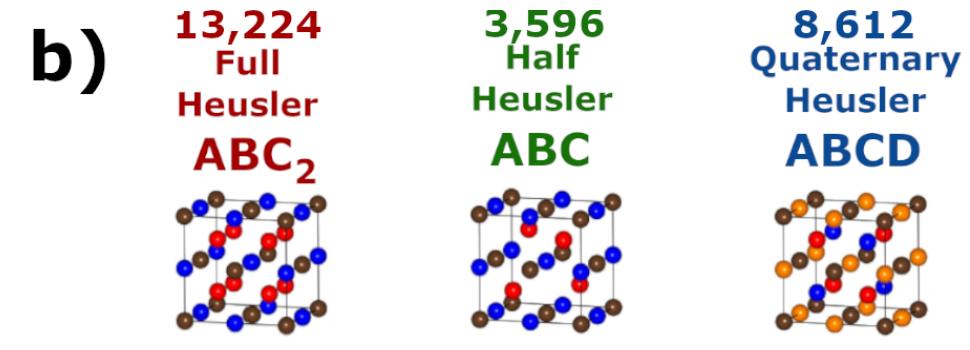
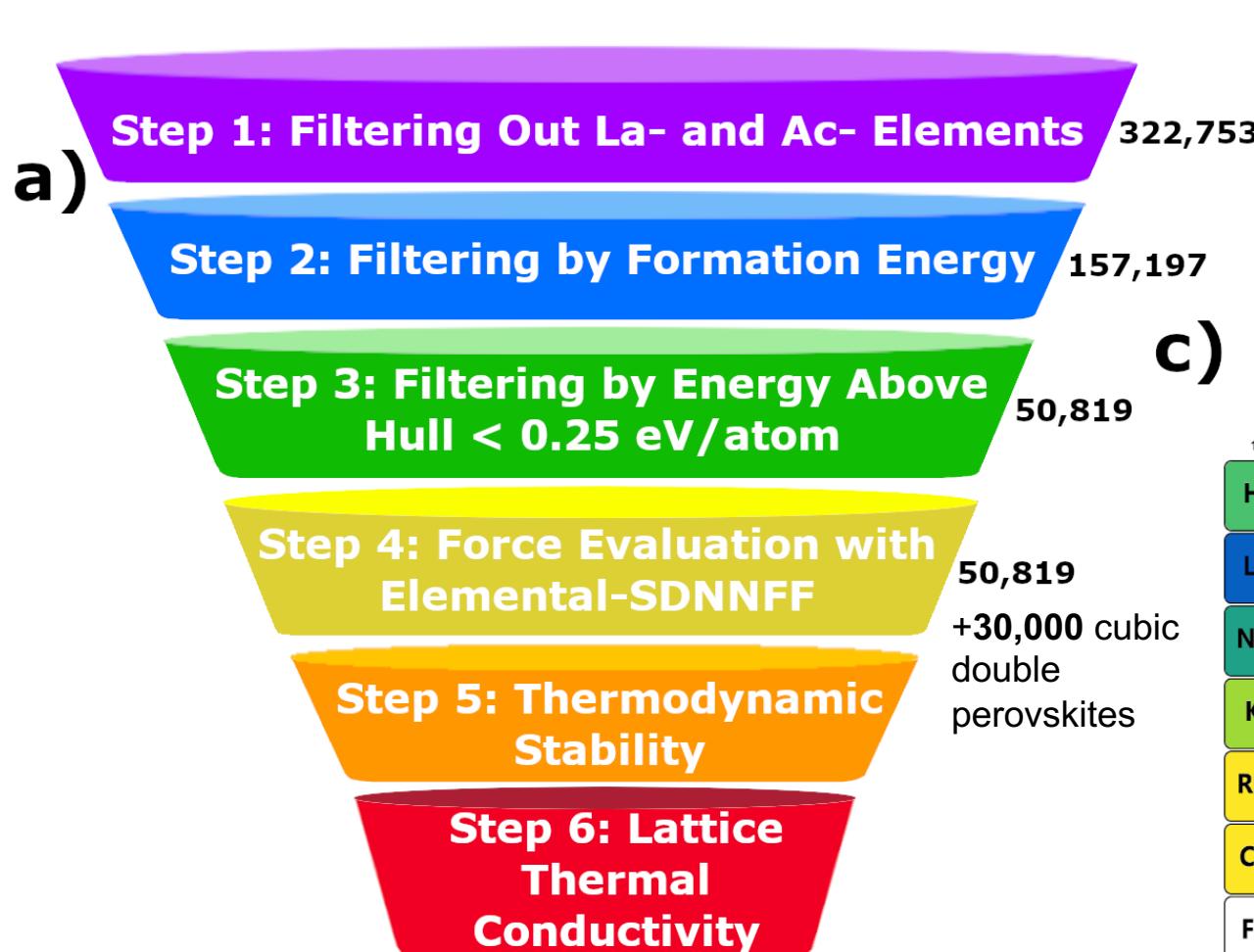
# ➤ Elemental-SDNNFF: Active Learning

*Active learning loop for model improvement with the query by committee method*



# ➤ Million-Scale Data Integrated Sing Deep Neural Network Training

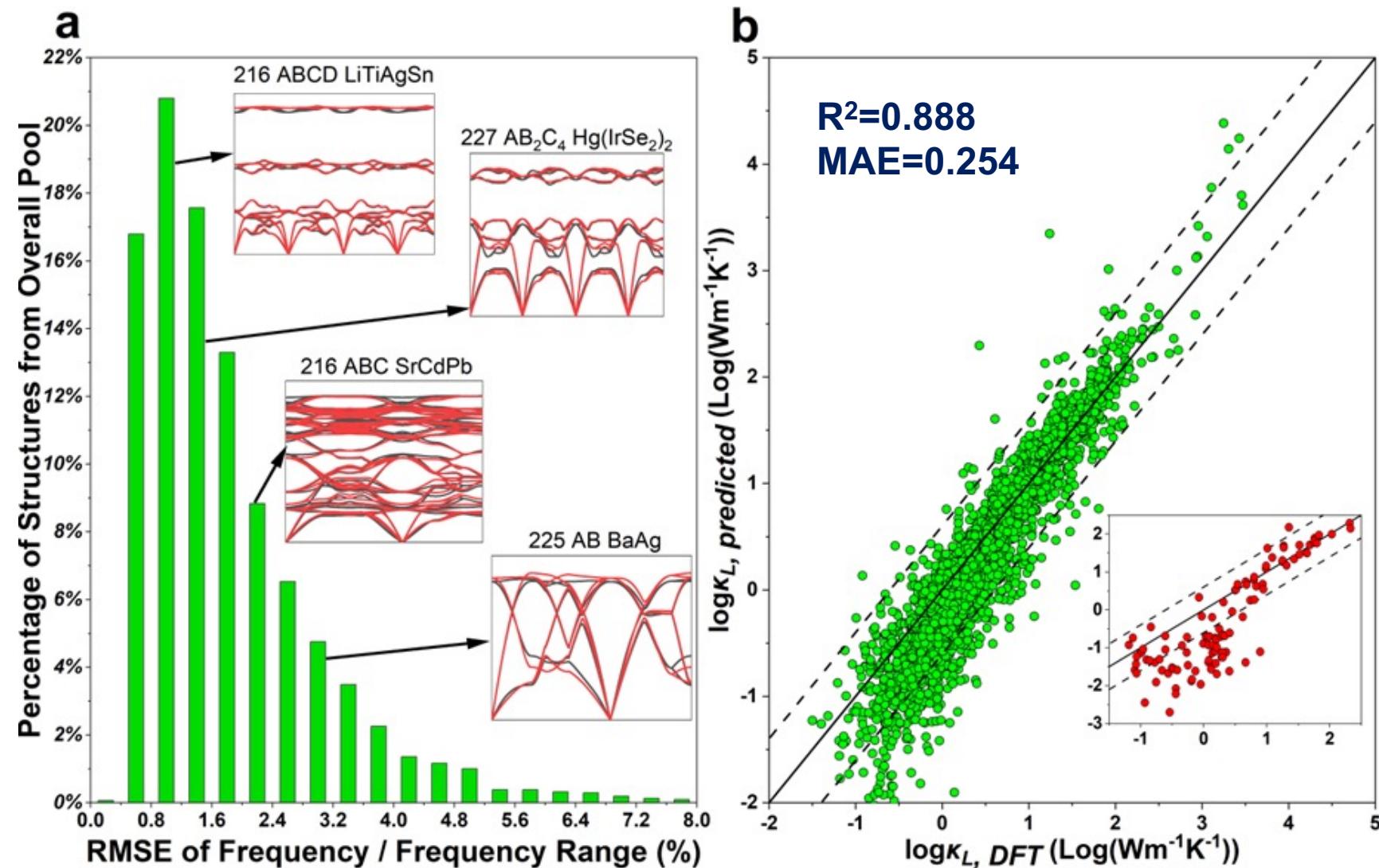
**~150,000** supercells →  **$1.2 \times 10^7$**  raw atomic configurations →  **$4 \times 10^7$**  after data augmentation



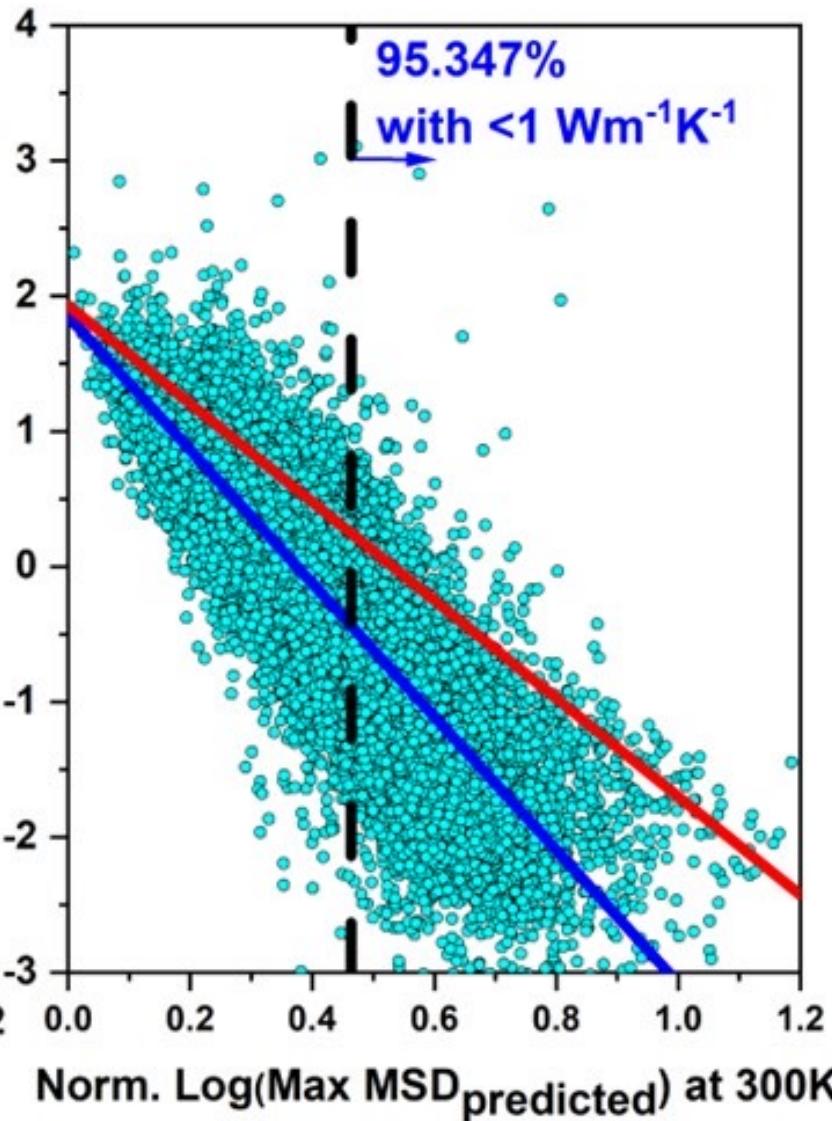
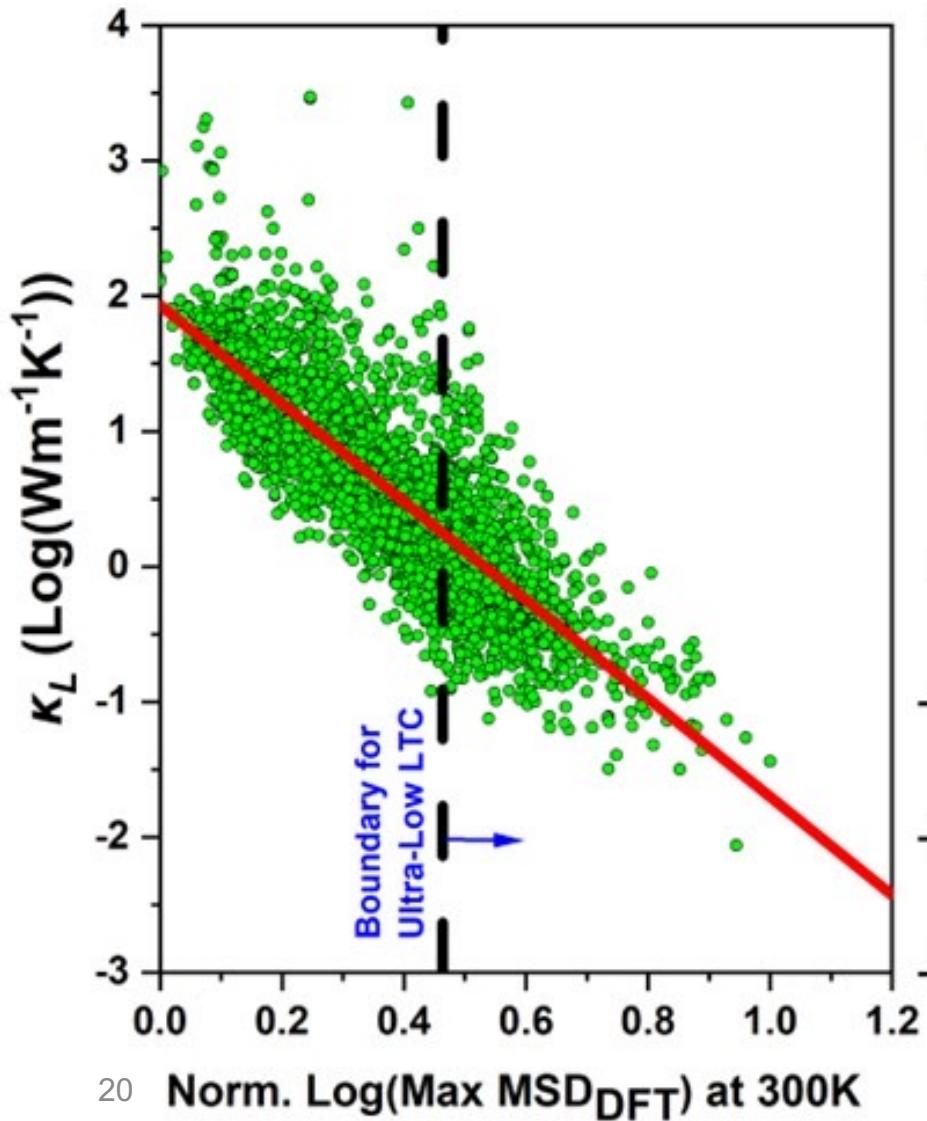
# ➤ New Deep Learning on ~80,000 Cubic Structures

Training on **3,107** cubic structures and predicting **~80,000** structures

- **4** space groups
- **17** types
- **63** elements



# ➤ Predicting Lattice Thermal Conductivity by Deep Learning



Mean Square Displacement (MSD)

$$\langle |u^\alpha(jl, t)|^2 \rangle = \frac{\hbar}{2Nm_j} \sum_{\mathbf{q}, \nu} \omega_\nu(\mathbf{q})^{-1} (1 + 2n_\nu(\mathbf{q}, T)) |e_\nu^\alpha(j, \mathbf{q})|^2.$$

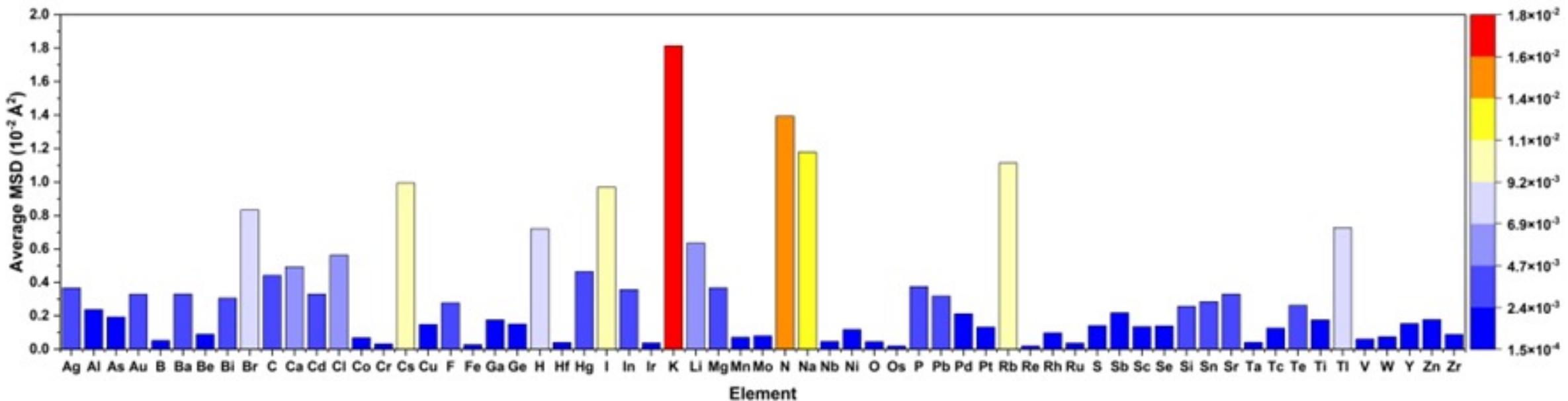
## Facts:

- **77,091** new cubic structures screened
- **27,059** (35%) structures have positive dispersions
- **8,873** structures have LTC below 1 W/mK



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# ➤ Insight into Ultralow Lattice Thermal Conductivity



- Majority of structures with large MSD values show ultralow LTC
- Alkali metal elements (Na, K, Rb, Cs) have large MSD and most of corresponding structures show strong phonon anharmonicity due to the **rattling effect**
- Mean Square Displacement (MSD) is a simple and ML-trainable descriptor or parameter that can be used for predicting low LTC materials

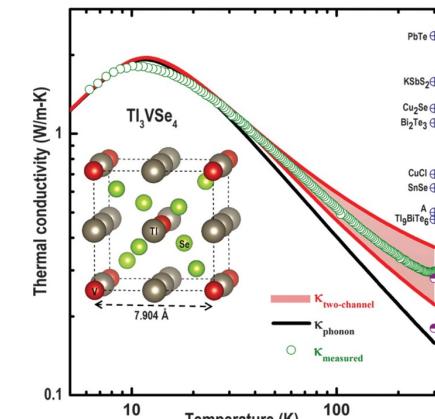
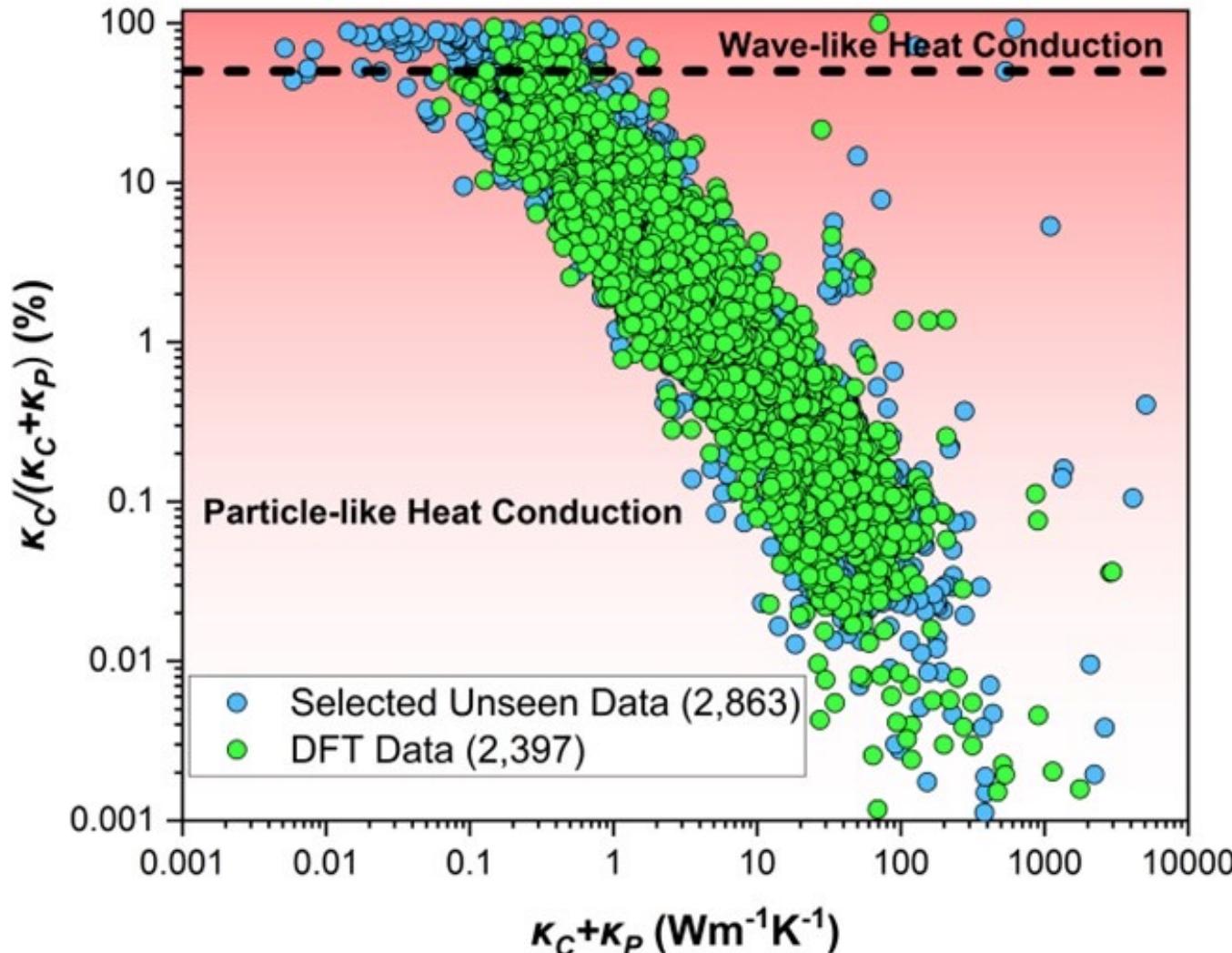


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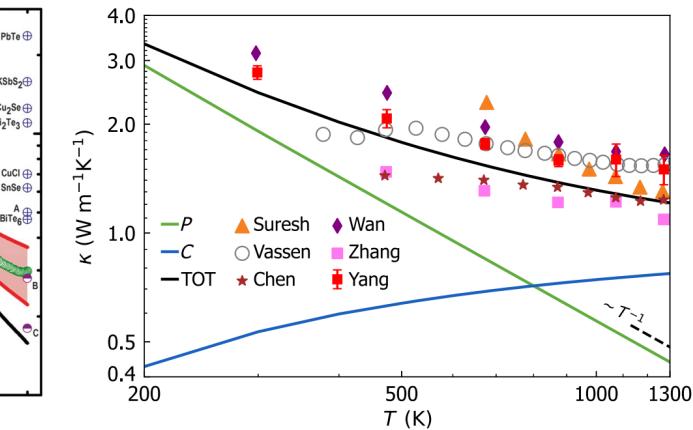
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# ➤ Two-Channel Thermal Transport (Off-Diagonal Contribution)



Science 360, 1455 (2018)

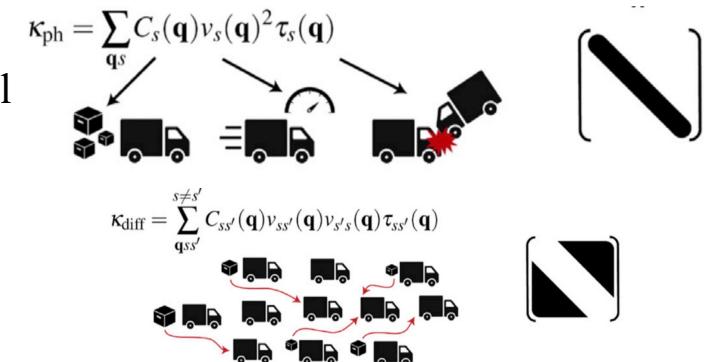


Phys. Rev. X 12, 041011 (2022)

Propagons or Diffusons?

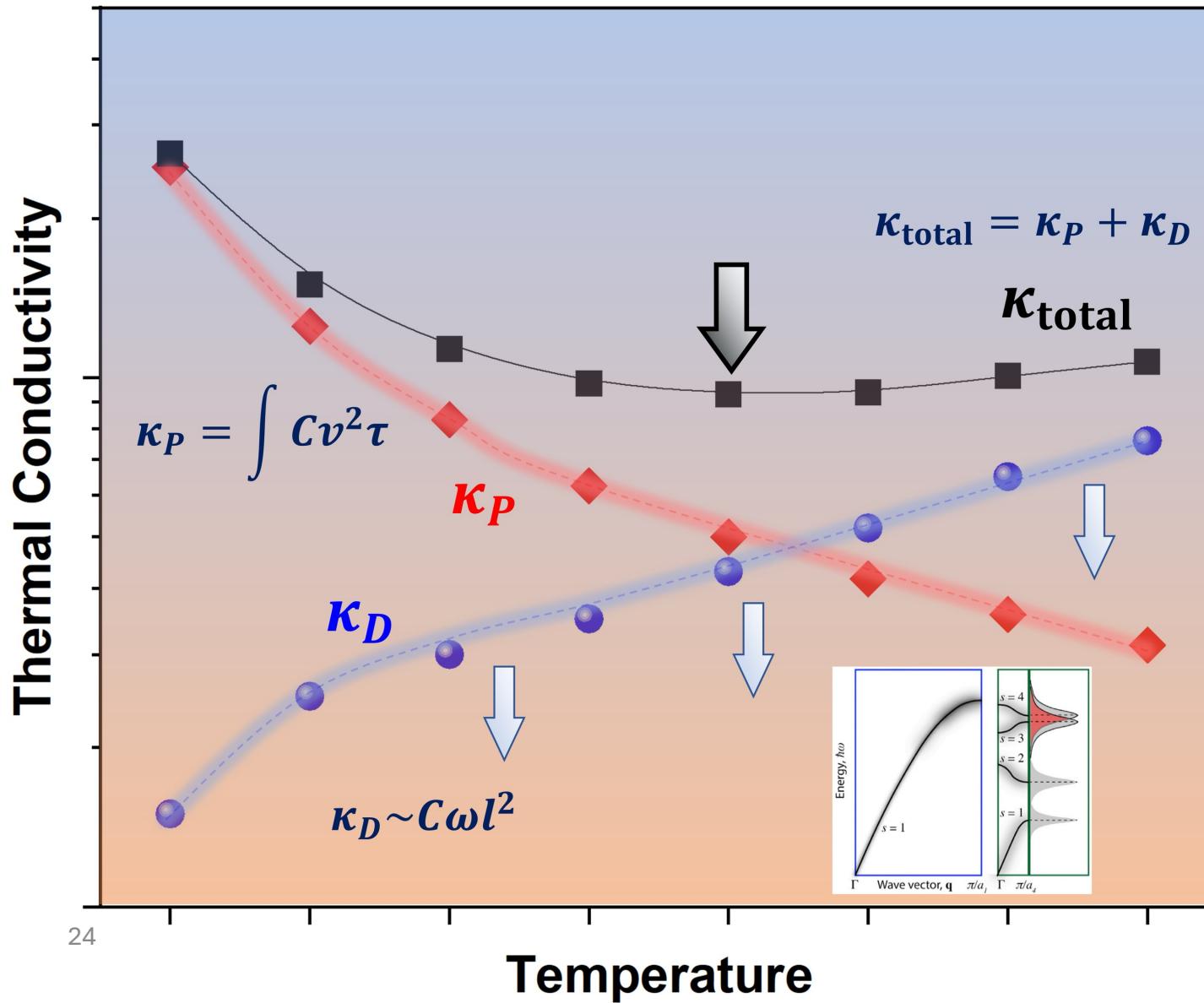
Phonon-gas channel

Diffuson channel:  
Off-diagonal  
contribution to  
thermal transport



Mat. Today Phys. 18, 100344 (2021)

# ➤ What is the Lower Limit of Lattice Thermal Conductivity?



125 QUESTIONS:  
EXPLORATION AND DISCOVERY



<https://www.science.org/content/resource/125-questions-exploration-and-discovery>

**What are the limits of heat transfer in matter?**

Materials science research may help us address the challenge of high power-density heat transfer, from nanometer-scale devices such as circuits to industrial-scale processes such as power plants.



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# ➤ High-Throughput DFT Data Generation

- Procedure for DFT-LTC calculation
  - Optimize the primitive cell
  - Screen for non-zero bandgap
  - Duplicate primitive cells and displace atoms from equilibrium position
  - Evaluate atomic forces in supercells by DFT
  - Perform IFCs fitting up to 3<sup>rd</sup> order by CSLD
  - Iteratively solve BTE for the diagonal term LTC ( $\kappa_p$ )
  - Compute off-diagonal component ( $\kappa_c$ )

$$\kappa_c^{\alpha\beta} = \frac{\hbar^2}{2k_B T^2 N\Omega} \sum_q \sum_{j,j'}^{j \neq j'} V_{j,j'}^\alpha(q) V_{j',j}^\beta(q) \times (\omega_j(q) + \omega_{j'}(q)) \times \frac{\omega_i(q)n_i(q)(n_i(q) + 1) + \omega_{i'}(q)n_{i'}(q)(n_{i'}(q) + 1)}{4(\omega_j(q) - \omega_{j'}(q))^2 + (\Gamma_j(q) + \Gamma_{j'}(q))^2} \times 2(\Gamma_j(q) + \Gamma_{j'}(q))$$

*Cross-Group Velocity*      *Cross-Heat Capacity*

*Cross-Lifetime*

- 1,212 total structures for our analysis

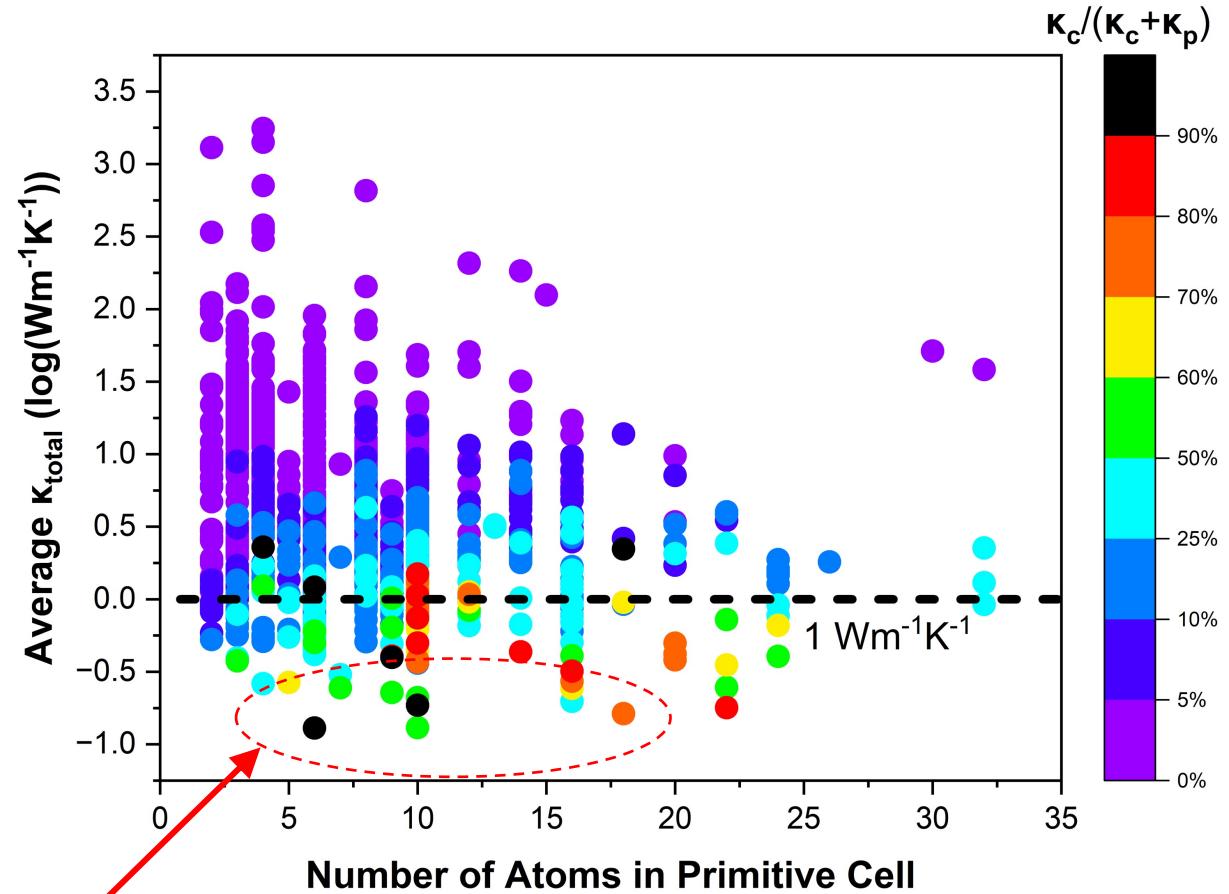


Our goal: searching materials with lowest possible total LTC ( $\kappa_{total} = \kappa_p + \kappa_c$ )



# ➤ Design Principles for Achieving Ultralow Total LTC

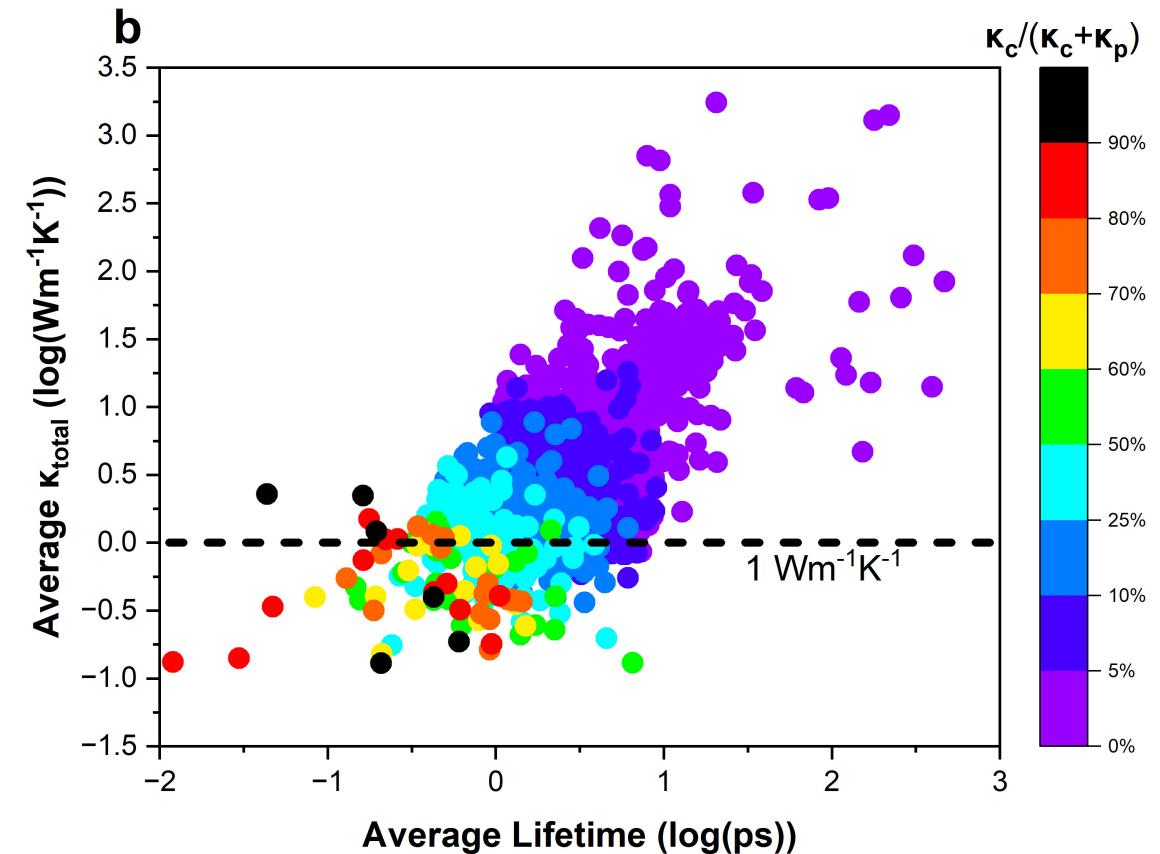
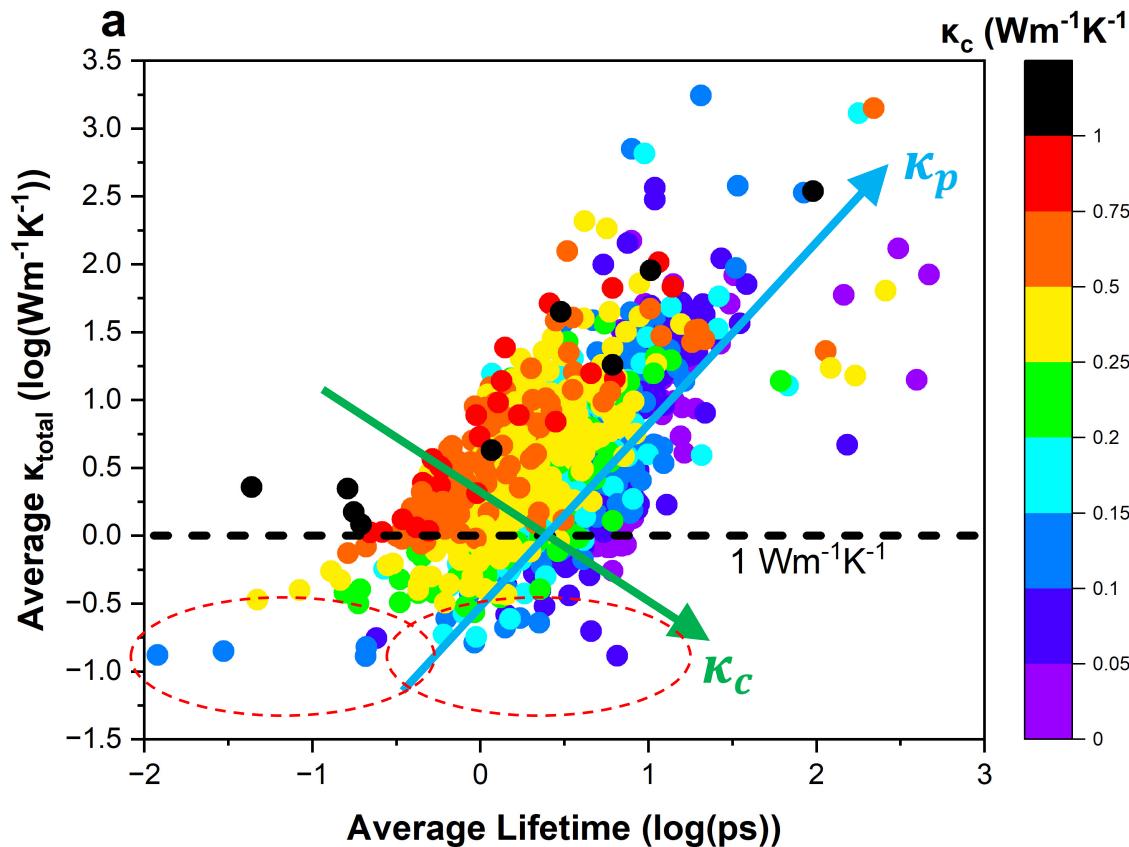
- Number of atoms in primitive cell vs. total  $\kappa$ 
  - Low atoms = high symmetry
  - Symmetry promotes  $\kappa_p$
  - High atoms (low symmetry) promotes  $\kappa_c$
- 90% of  $\kappa < 1 \text{ Wm}^{-1}\text{K}^{-1}$  exists in the <16 atom range
- Formula for low  $\kappa$ :
  - Medium to low number of atoms (low  $\kappa_c$ )
  - High anharmonicity (low  $\kappa_p$ )
    - Rattling atoms
    - Lone-pair electrons / resonant bonding
    - Meta-valent bonding
    - Bond hierarchy
    - .....



Promising searching area: simple  
to medium complex crystals?



## ➤ Two Scenarios for Extremely Low $\kappa_{total}$



(a) Lifetime is extremely short: both  $\kappa_c$  and  $\kappa_p$  are low

(b) Lifetime is intermediate:  $\kappa_c$  and  $\kappa_p$  are comparably low

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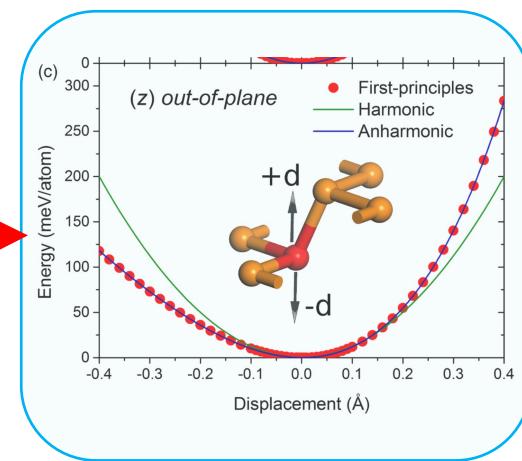
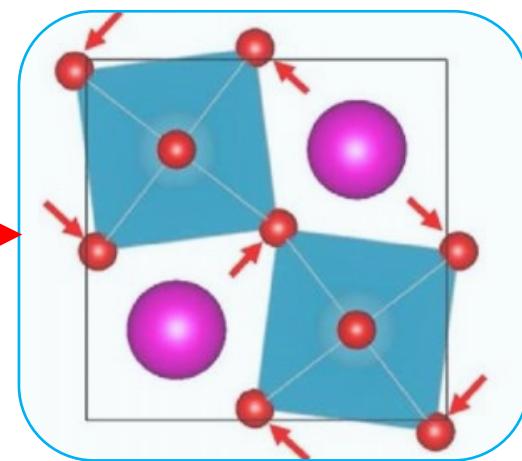
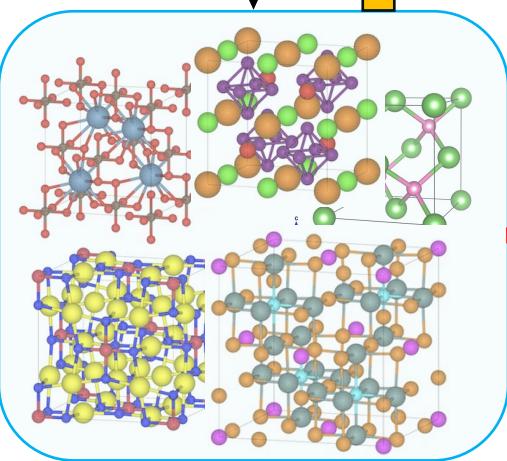
**Generate Hypothetical Structures**

**Big issue: limited data & lack of comprehensive properties**

**Direct Method**

**AI/ML Training & Forward Prediction**

**Phonon Database**



- Phonon Dispersions  
• Thermal Conductivity  
• Relaxation Time  
• Heat Capacity  
• Scattering Channels  
• Two-Channel Transport  
• Phonon Hydrodynamics  
• Mechanical Stability  
• .....

Comprehensive Phonon Properties

Indirect Method (Elemental-SDNNFF)

**Inverse Design**

More Complex Phonon Mediated Properties

- Superconductors
- Photovoltaics
- Superionic Conductors
- Phase Change
- .....

Beyond phonons



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# THANKS!

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<https://scholar.google.com/citations?user=8Wt2yS0AAAAJ&hl=en>



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