MRS Fall EQ04 Tutorial: Predicting phonon DoS with e3nn

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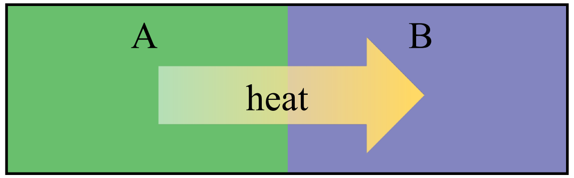
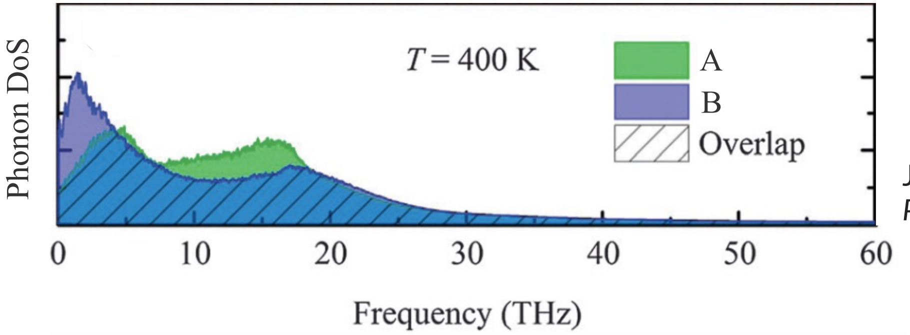
Motivation | Phonon DoS and thermal properties

- Phonon density of states (DoS): $g(\omega) = \frac{1}{3nN}\sum_{\mathbf{k}\nu}\delta(\omega - \omega_{\mathbf{k}\nu})$
- Lattice heat capacity: $C_V = 3nNk_B \int_0^{\omega_L} \left(\frac{\hbar\omega}{2k_BT}\right)^2 \operatorname{csch}^2\left(\frac{\hbar\omega}{2k_BT}\right) g(\omega)d\omega$
- Thermal conductivity: $k = \frac{1}{3}nC_V\langle v\rangle\lambda$
- Interfacial thermal transport: $E_{prop} = \int \omega g_{\cap}(\omega)\, d\omega$





J. Zhang, et al. *Physical Chemistry Chemical Physics* 17.37 (2015): 23704-23710.

- Phonon-mediated superconductivity: $\lambda = 2\int_0^\infty d\omega\, \alpha^2(\omega)\, g(\omega)/\omega$

$$\overline{\omega} = \exp\left(\frac{2}{\lambda}\int_0^\infty d\omega\, \ln(\omega)\alpha^2(\omega)\, g(\omega)/\omega\right)$$

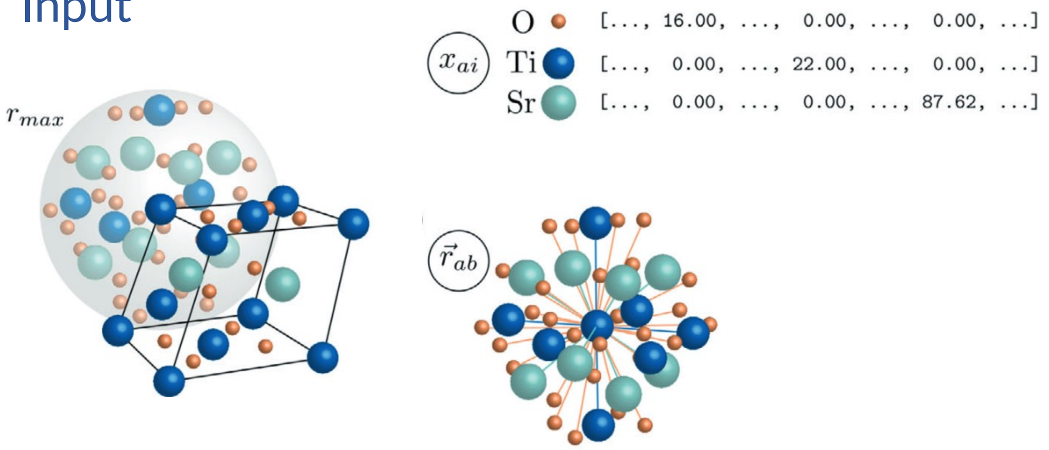
$$T_c = \frac{\overline{\omega}}{1.2}\exp\left(\frac{-1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right)$$

Motivation | Challenging to obtain phonon DoS

- Experimental
  - Inelastic neutron and X-ray scattering at scientific user facilities
- Computational
  - *Ab initio* calculations scale with $O(N^4)$, N = number of atoms in the calculated structure
  - High computational cost of disordered or highly complex systems
- Data-driven
  - Limited training data ($\sim 10^3$)
  - High-dimensional output depending on resolution
  - Data augmentation to capture arbitrary rotations of crystal structures
- Euclidean neural networks (e3nn)
  - Symmetry-aware --> no data augmentation
  - Equivariant to 3D translations, rotations, and inversion --> constrain function optimization space
  - Extend naturally to systems with substitutional disorder

Approach | Predicting phonon DoS with e3nn

Input



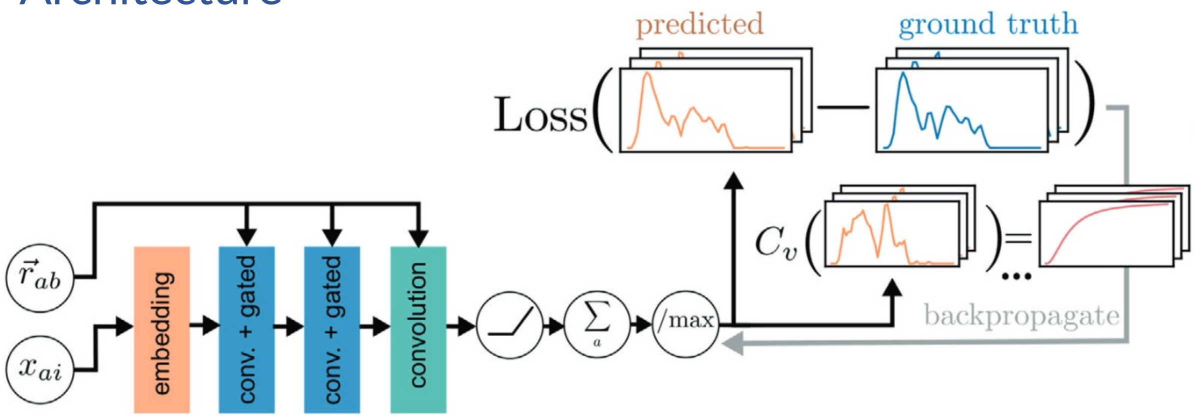
- Build graphs from crystal structures:

  Each atom becomes a node with a feature x_{ai} expressed in the irrep "118x0e"

  Atomic mass-weighted one-hot encoding: 118 scalars with atomic mass at the Z-th scalar (zero otherwise)

  Edge $\overrightarrow{r_{ab}}$ is formed between pairs of atoms with $r \leq r_{\max}$

Architecture



- Manipulations of irreps within the neural network:

  Embedding, convolution, gated block (activation)

- Interpretation and results:

  Visualization of intermediate learned features

  Predict partial DoS which NN is not explicitly trained to do

  Predict phonon DoS of substitutional alloys with N-hot input

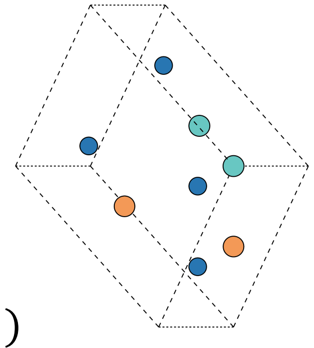
Z. Chen, N. Andrejevic, T. Smidt, et al. *Advanced Science* (2021): 2004214.

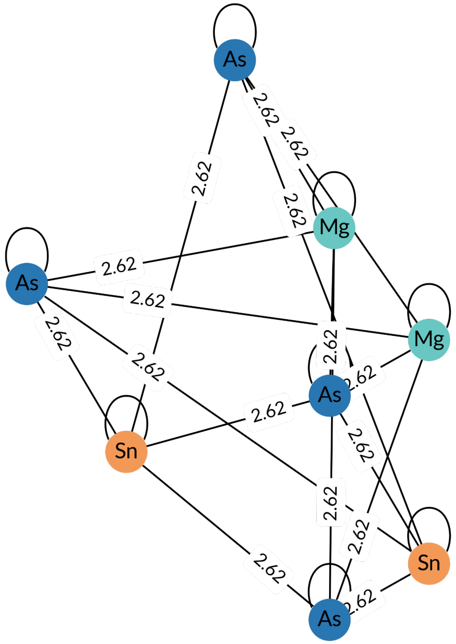
Approach | Some considerations

- Periodicity

  Adding periodicity only changes the graph, not the model

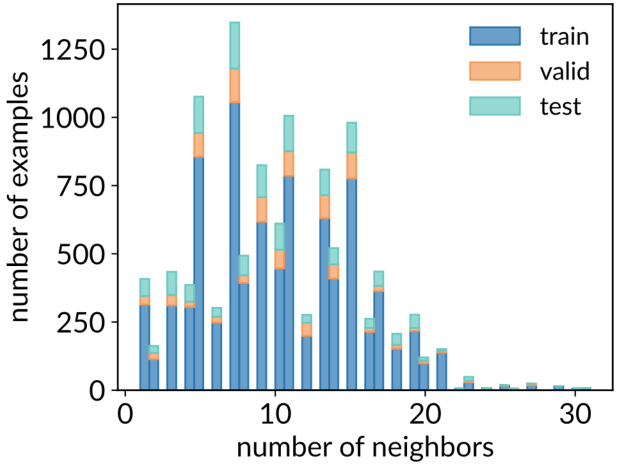
  Edges can be formed between an atom and its images (if $r \leq r_{\max}$)





- Normalization

  Average number of neighbors z: $f'_a = \frac{1}{\sqrt{z}} \sum_{b \in \partial(a)} f_b \otimes_{|\boldsymbol{r}_{ab}|} Y(\boldsymbol{r}_{ab}/|\boldsymbol{r}_{ab}|)$



- Generalizability

  Weighting one-hot features by atomic mass allows the model to generalize better to unseen elements (over simple one-hot encoding)