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## Motivation

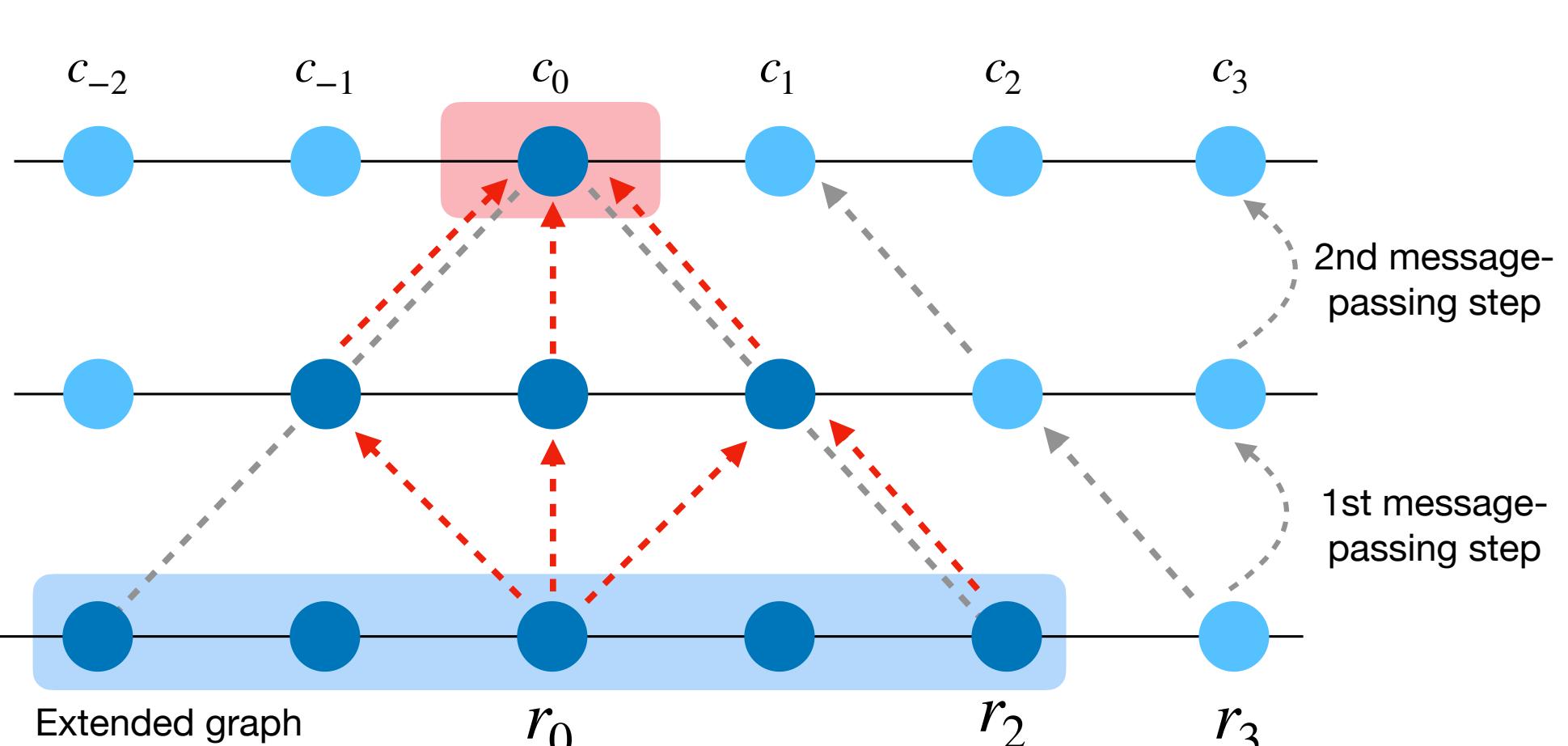
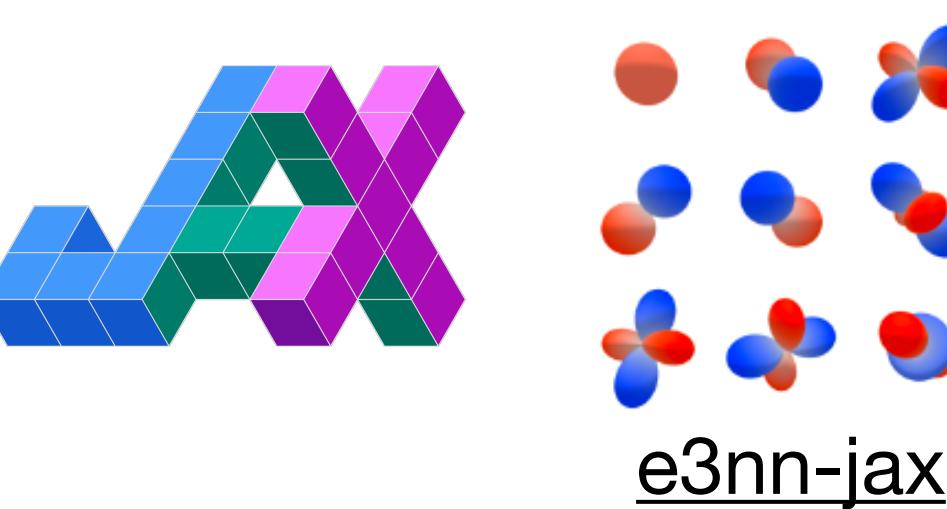
Phonons are crucial in many physical properties and applications

- Thermoelectric and phonon transport
- BCS-type superconductors
- Structural stability and soft modes
- Atomic vibrations governed by the dynamical matrix

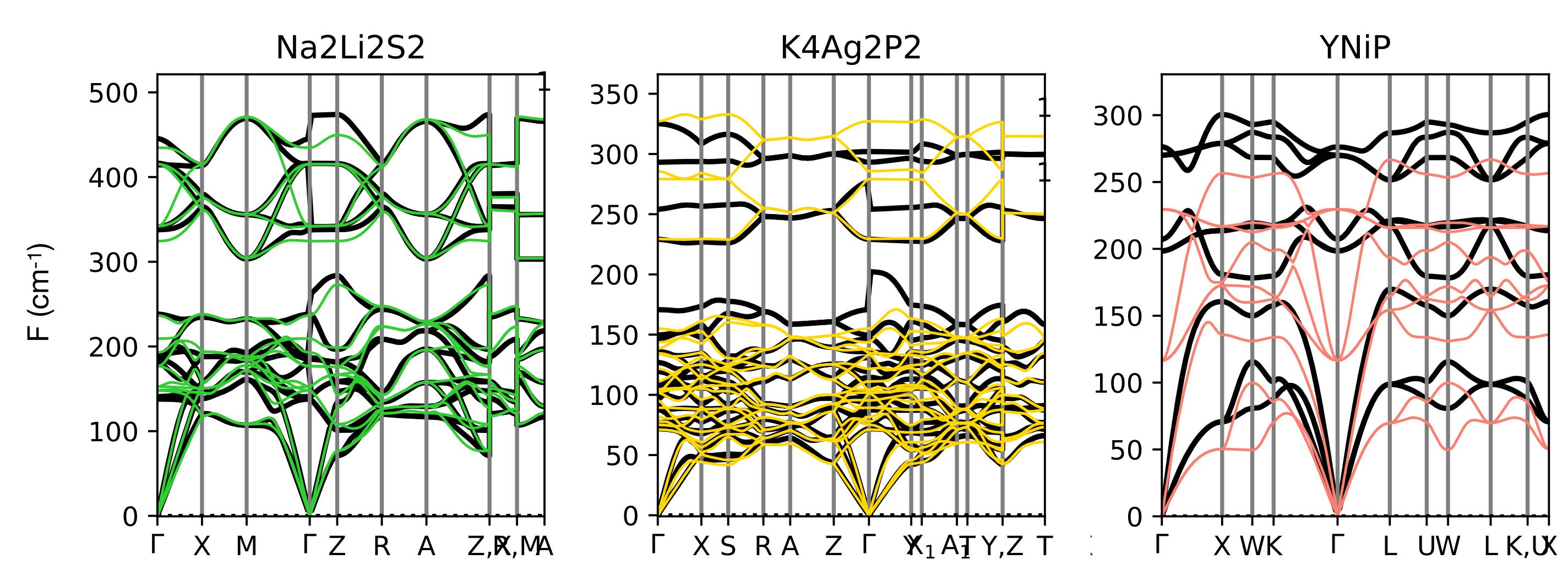
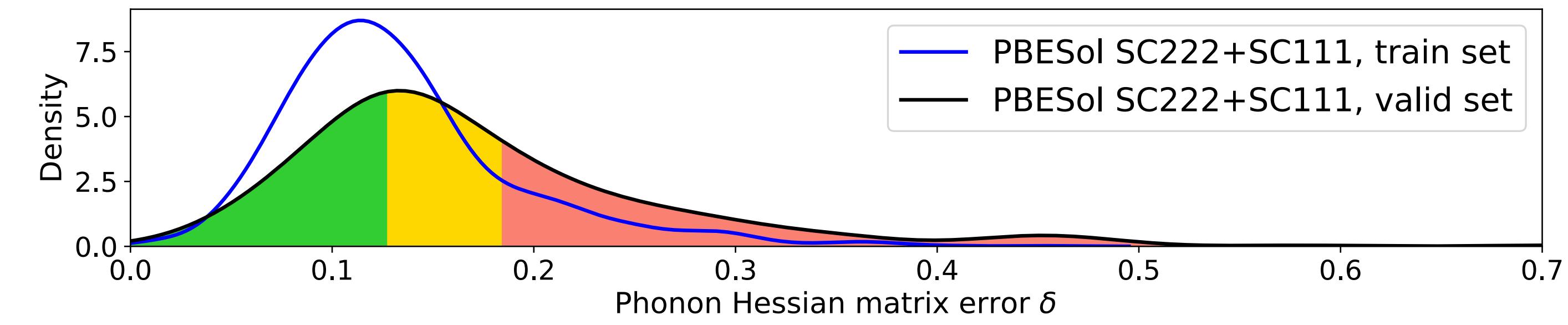
$$D_{\alpha\beta}(ij, \vec{q}) = \frac{1}{\sqrt{m_i m_j}} \sum_a \frac{\partial^2 E}{\partial(\vec{x}_{0i})_\alpha \partial(\vec{x}_{0j})_\beta} e^{i\vec{q} \cdot (\vec{x}_{aj} - \vec{x}_{0i})}$$

## Phonax framework

- Energy model with equivariant NN (NequIP, MACE)
- e3nn-jax library and Hessians from JAX Autodiff
- Extended graph construction for periodic crystals



## Energy model based phonon predictions for periodic crystalline solids

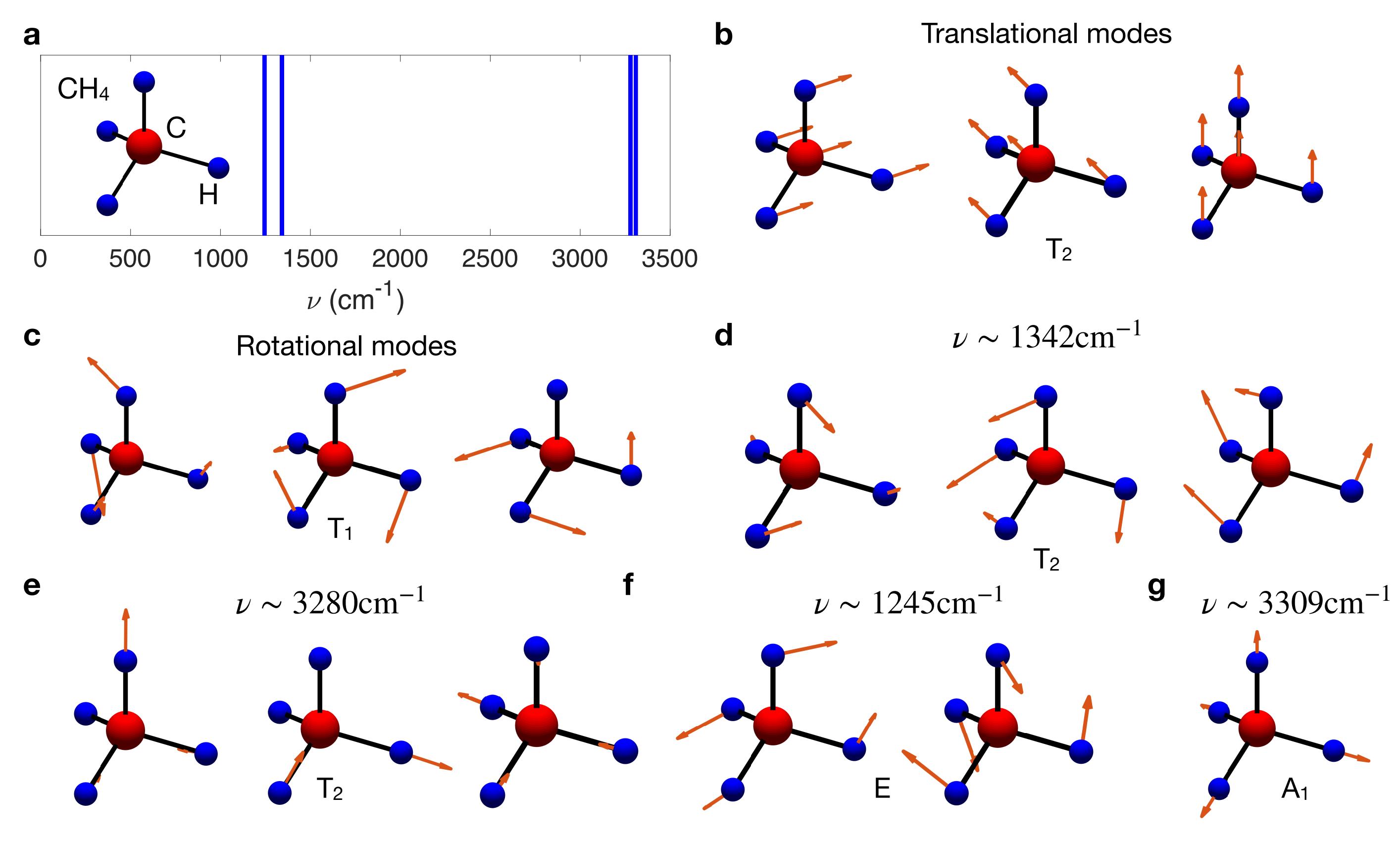


- NequIP Energy model trained with 1.2k inorganic crystals (PBESol VASP DFT)

## Symmetry irreps and IR / Raman modes from molecular Hessians

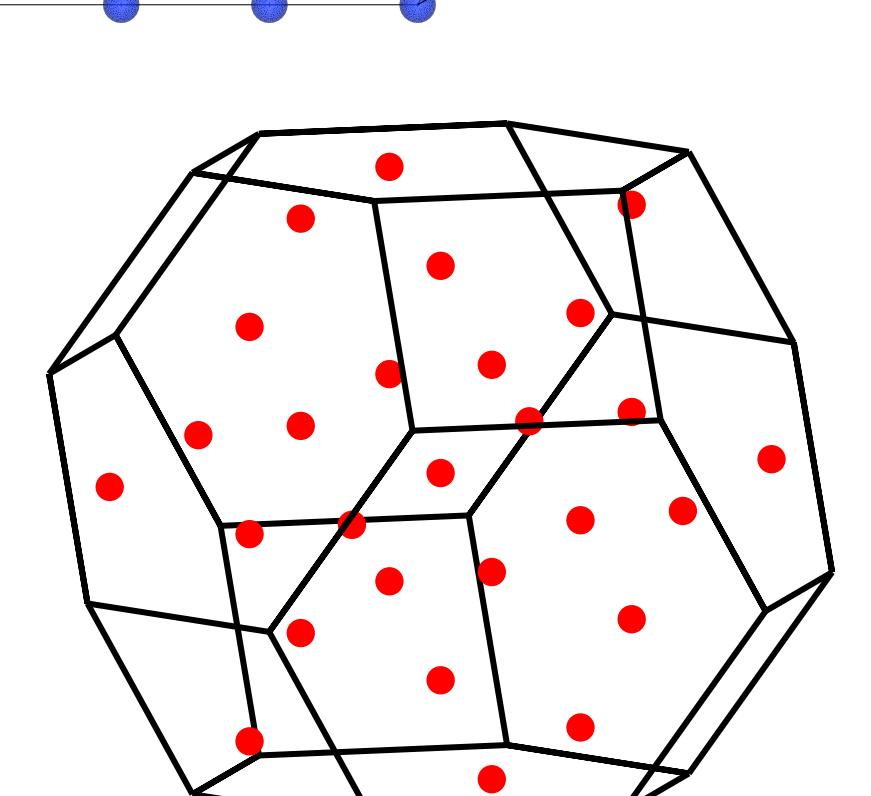
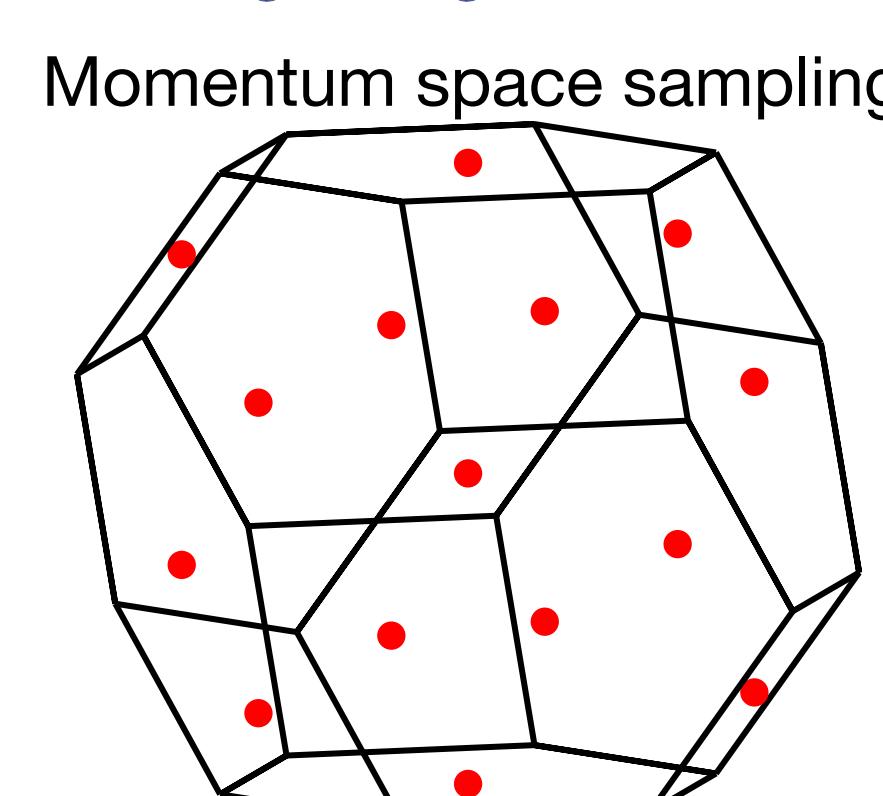
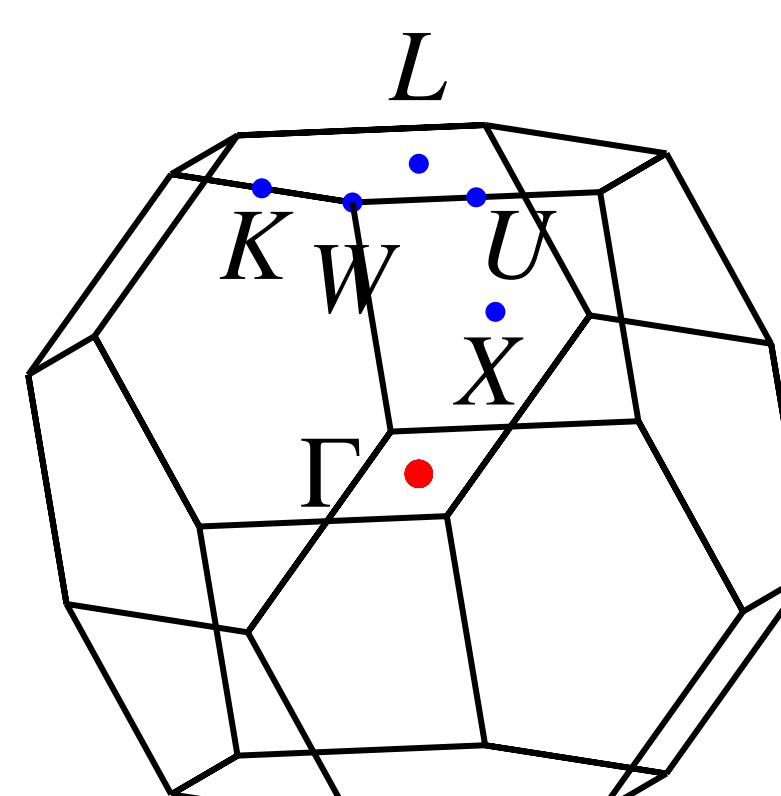
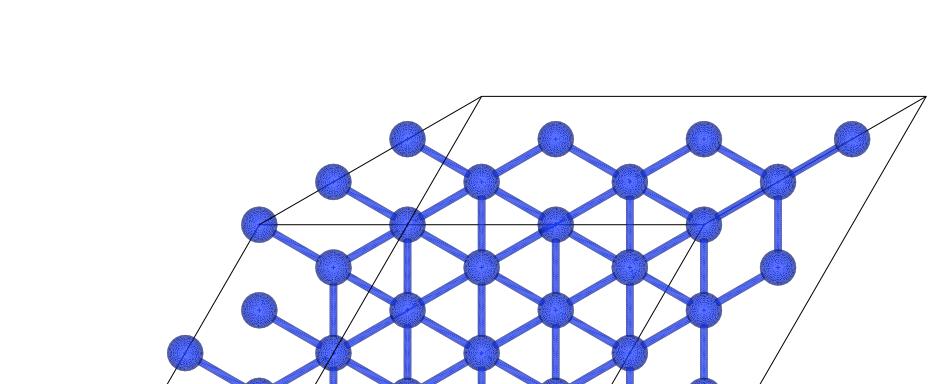
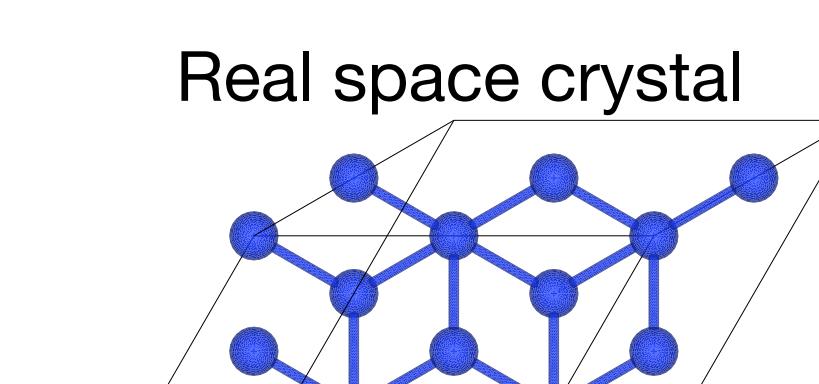
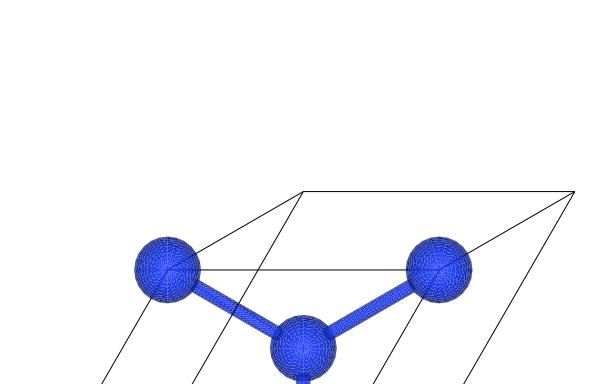
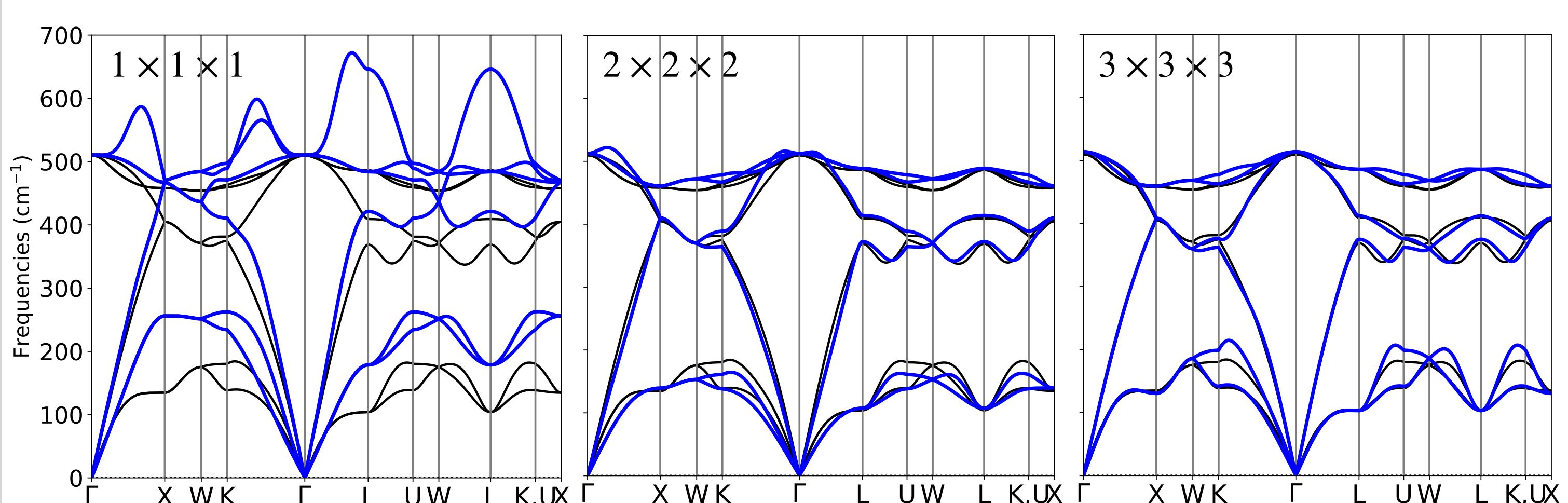
- IR / Raman spectroscopy and neutron scattering to identify molecules
- IR / Raman active modes from symmetry selection rules
- Derivations for the vibrational modes and their symmetry irreps

Ex: CH<sub>4</sub> molecule ( $T_d$  point group) and the IR active ( $T_2$ ) / Raman active ( $A_1+E+T_2$ ) modes



Ref: posym library for processing point group symmetries

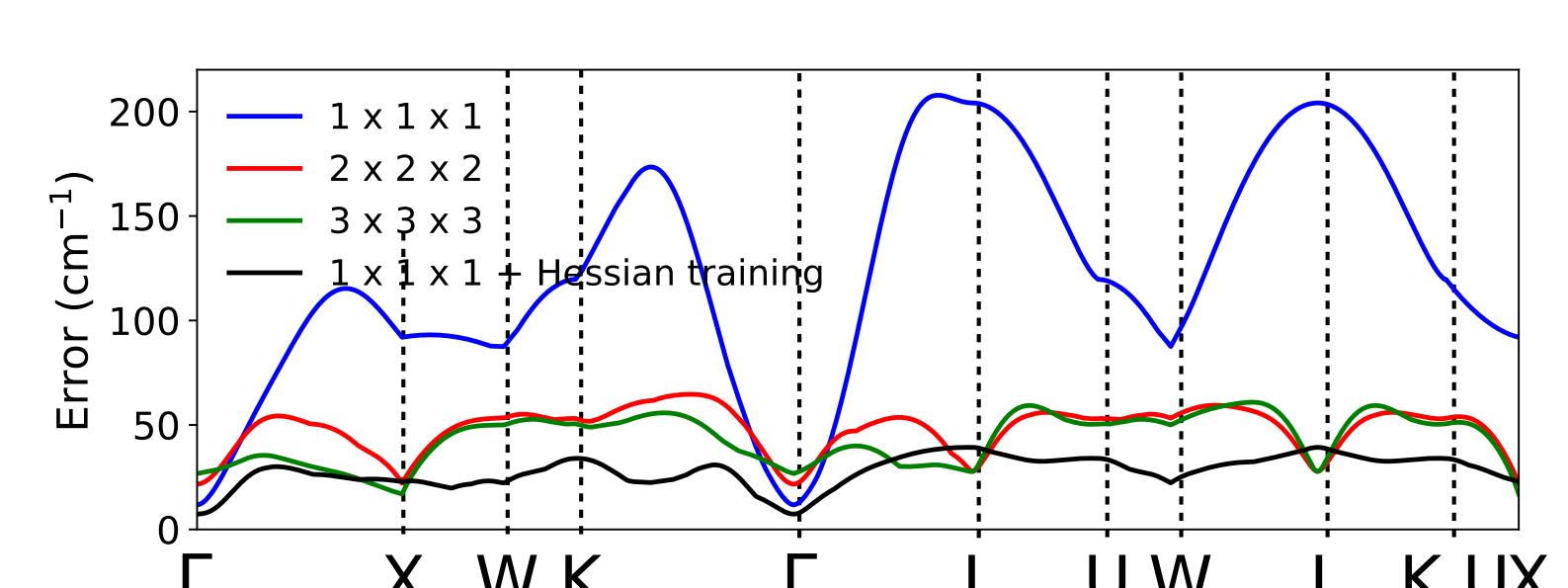
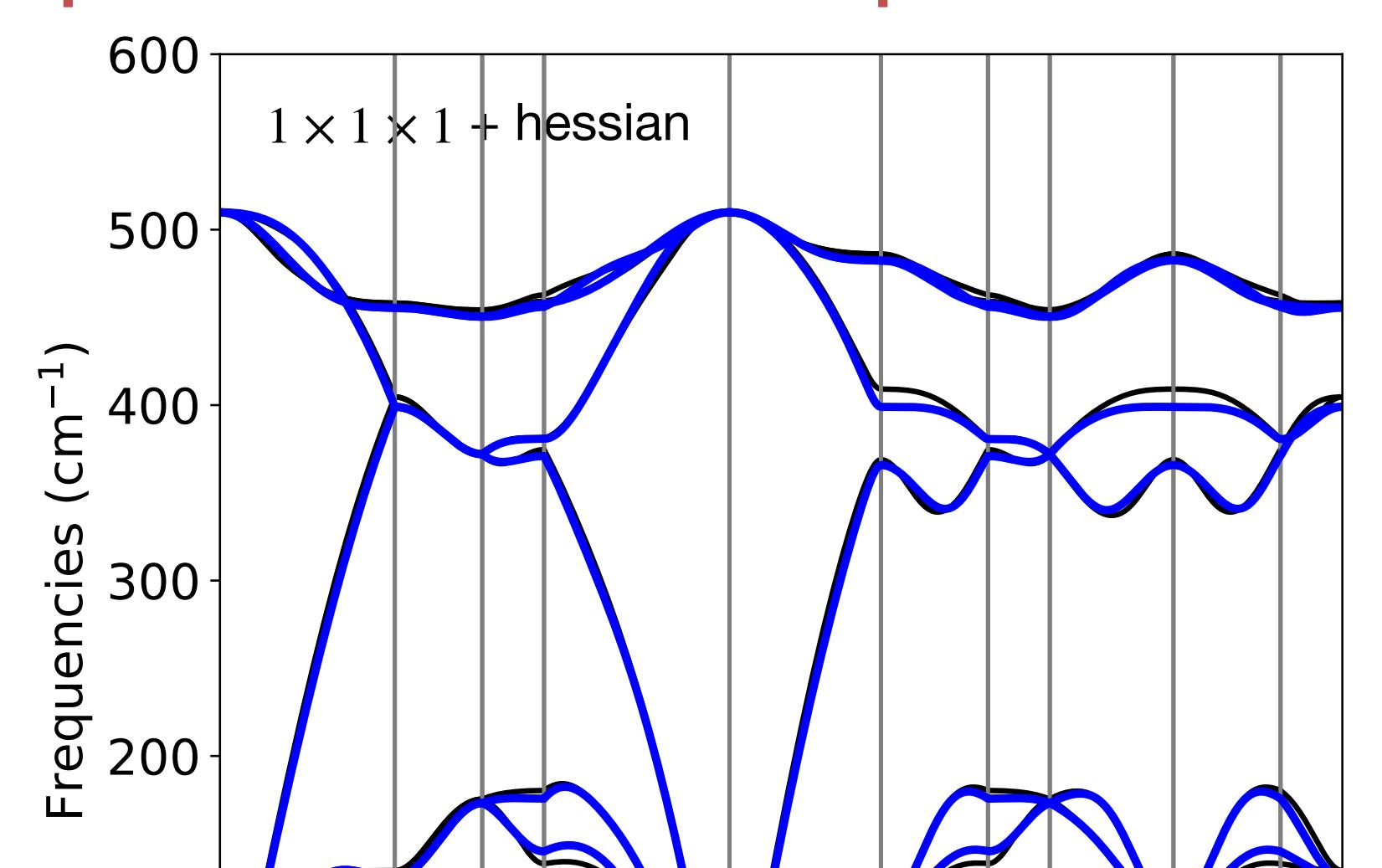
## Supercell energy / force data needed for better phonon predictions



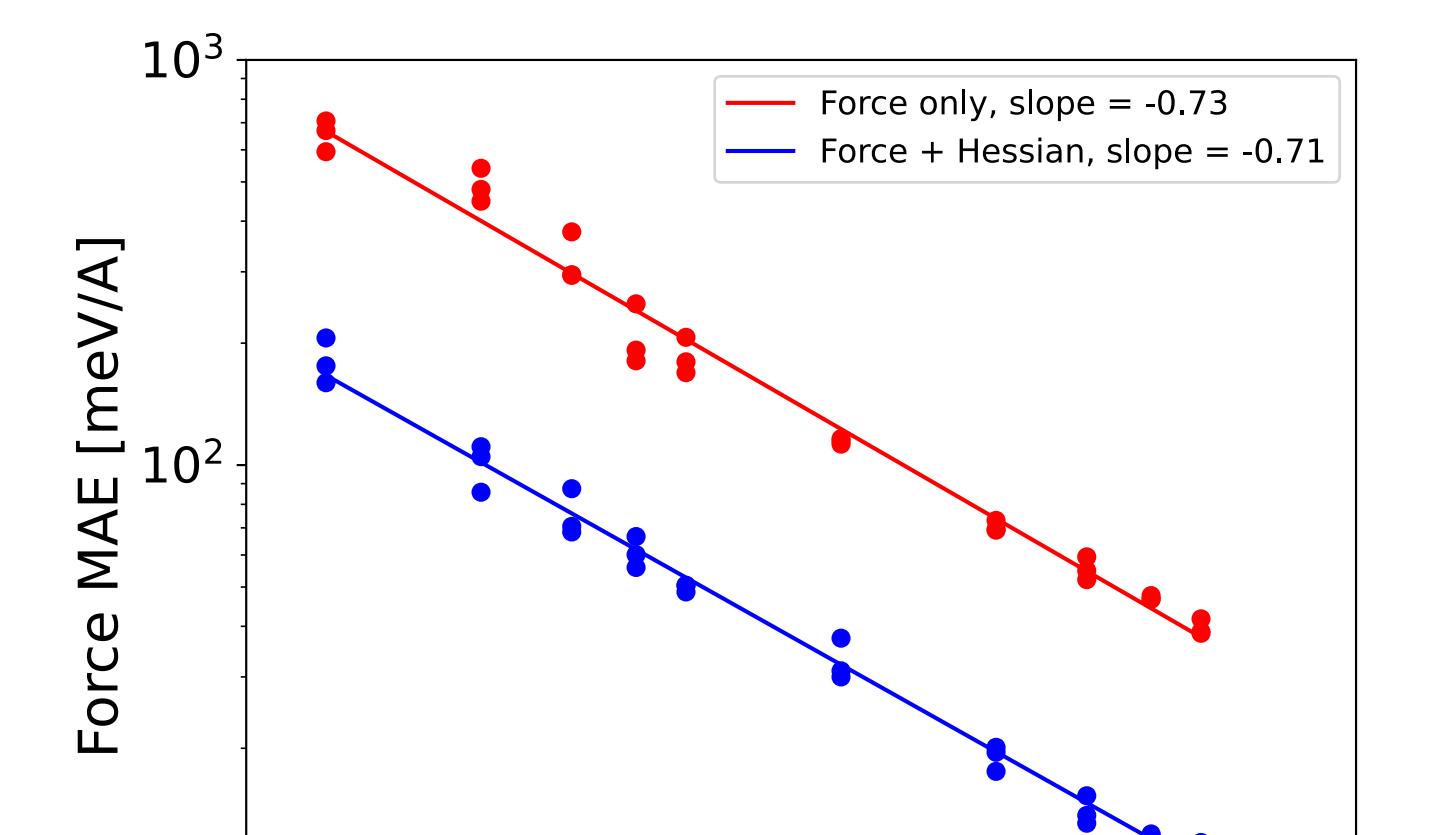
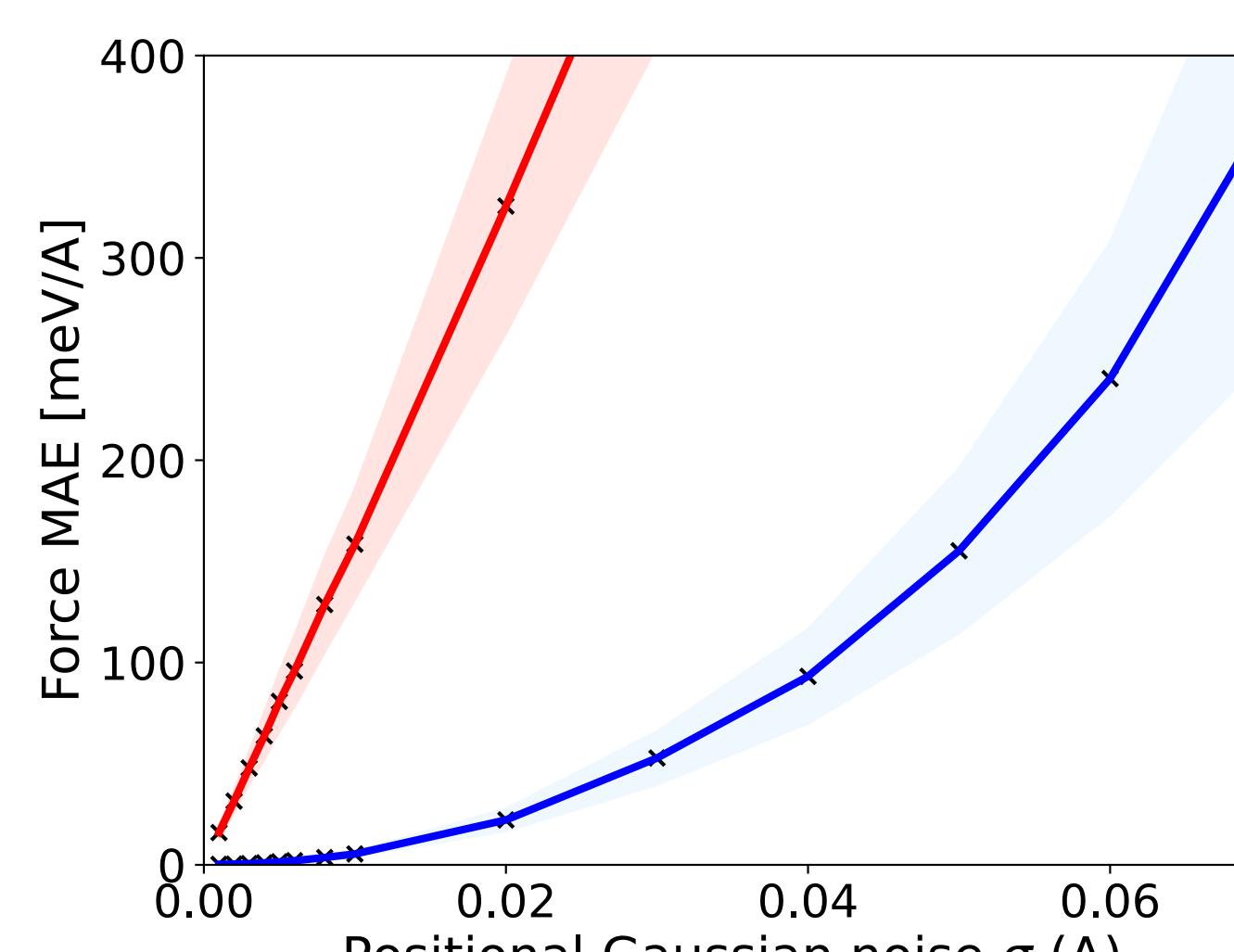
## Hessian data improves phonon predictions without supercell data

Energy model training:

- Supercell energy/force data needed for good phonon spectrum prediction
- Equivalently, finer k-point sampling in the momentum space
- Second derivative Hessians can be used as higher order training data to improve energy model without supercell energy/force data.



## Improving energy model force predictions with molecular Hessians



- Molecular Hessian strongly improves the local energy landscape and can be combined with force data to improve the training
- Hessian data effectively augments the training dataset size (NequIP)
- (Open question): Would training with Hessian data lead to better and more stable molecular dynamics simulations?

## References

- e3nn: Euclidean Neural Networks, Mario Geiger and Tess Smidt, arXiv:2207.09453
- E(3)-equivariant graph neural networks for data-efficient and accurate interatomic potentials, Simon Batzner et al., Nature Communications 13, 2453 (2022)
- MACE: Higher Order Equivariant Message Passing Neural Networks for Fast and Accurate Force Fields, Ilyes Batatia et al., arXiv:2206.07697