

# Transferable Machine-Learned Potentials for Vibrational Dynamics from Acene Crystals to Single-Molecule Host–Guest Systems

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Vibrational interactions in molecular crystals govern energy transport, decoherence, and anharmonic relaxation processes that are central to organic electronics and emerging molecular quantum platforms. Yet, accurately describing these effects in molecular crystals at experimentally relevant length and time scales remains computationally prohibitive for first-principles methods, due to the combined challenges of their soft vibrational modes, anharmonicity, and long-range van der Waals (vdW) interactions.

Here we present a systematically constructed machine learning interatomic potential (MLIP) framework based on the equivariant MACE architecture [1] and committee-driven active learning [2] that enables accurate and scalable simulations of vibrational dynamics in polyacene molecular crystals spanning naphthalene to pentacene. Beyond conventional energy and force benchmarking, we propagate committee uncertainty into vibrational observables, including vibrational densities of states and anharmonic line broadening, thereby quantifying training-induced errors. Furthermore, we rigorously assess the interpolation and extrapolation capability of the MLIP by deploying distinct active-learning strategies across multiple acene members (see Figure 1), enabling the construction of a unified and transferable potential across chemically related crystals [3].

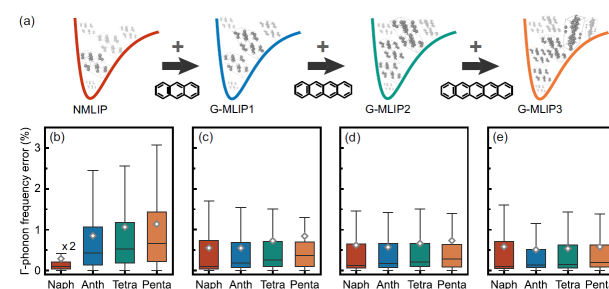
Importantly, we demonstrate that this generalized MLIP extends to large-scale host–guest systems while maintaining predictive accuracy (see Figure 2), enabling detailed simulations of vibrational coupling and decoherence effects relevant to molecular quantum technologies [4]. Our results further show that explicitly including van der Waals interactions is important for the generalized potentials to accurately capture long-range intermolecular forces. Large-scale molecular-dynamics simulations further reveal mode-selective hybridization between guest vibrations and host phonon manifolds and quantify environment-induced vibrational renormalization, providing insights for understanding and controlling vibrational relaxation in these systems.

Together, these results establish an uncertainty-aware and transferable framework for resolving anharmonic nuclear dynamics in complex molecular solids and provide a pathway toward rational engineering of vibrational interactions in molecular environments relevant to quantum technologies [5].

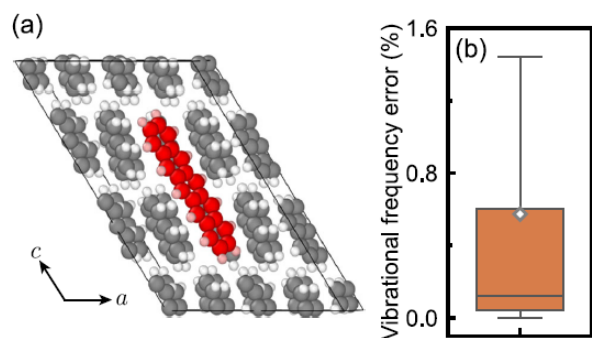
## References

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## Figures



**Figure 1.** (a) Sketch illustrating the active-learning scheme used to create generalized MLIPs. (b–e) Error box plots for the  $\Gamma$ -point phonon frequencies of naphthalene (Naph), anthracene (Anth), tetracene (Tetra) and pentacene (Penta) molecular crystals, predicted by N-MLIP (b), G-MLIP1 (c), G-MLIP2 (d) and G-MLIP3 (e). (Arial 9)



**Figure 2.** (a) Crystal structure of pentacene-doped  $2 \times 2 \times 3$  naphthalene molecular crystal, with  $a$  and  $c$  representing the crystal axes in monoclinic symmetry. Two naphthalene molecules along the front-facing plane are removed for illustration purposes. b Box plot showing the prediction errors for vibrational frequencies of the host-guest system.