Machine Learning-Driven Hamiltonian Matrix Prediction: Equivariant vs. Non-Equivariant Models

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We present HForge, a machine learning (ML) framework for predicting the Hamiltonian (H) and Overlap (S) matrices directly from atomic structures. Our approach employs descriptors extracted from MACE and utilizes Siesta for computing reference Hamiltonians, using amorphous boron nitride (aBN) as a test case. We investigate the performance of equivariant and non-equivariant ML models, assessing their ability to generalize from smaller training structures to larger systems—a key challenge given that training costs are 3-4 times higher than inference. Our results show that equivariant models achieve superior transferability by preserving symmetries inherent to quantum mechanical interactions, while non-equivariant models struggle with extrapolation. This study builds upon recent advancements in graph-based quantum representations [1] and universal graph-to-matrix conversions for electronic structure prediction [2], demonstrating the impact of graph-based descriptors on Hamiltonian learning. Our findings highlight the potential of ML-based Hamiltonian prediction for accelerating electronic structure calculations and enabling scalable quantum simulations.

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Figures

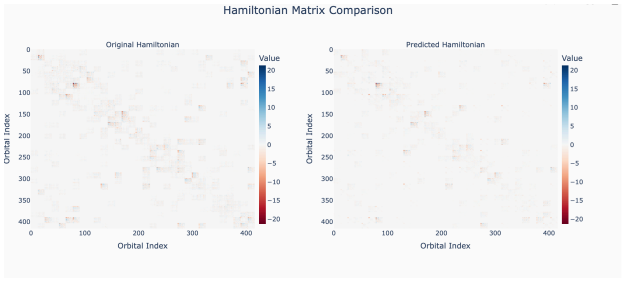

Figure 1. Comparison between DFT and ML Hamiltonian for BN structure (scale is in eV)

References

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