Machine Learning-Driven Hamiltonian Matrix Prediction Applied to aBN and hBN

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Andrei Voicu Tomut (1)
Stephan Roche(1,2) , Jose Hugo Garcia Aguilar(1)

1.Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and The Barcelona Institute of Science and Technology, Campus UAB, 08193 Bellaterra, Catalonia, Spain.
2.ICREA — Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain
)

Contact@E-mail (Century Gothic 10)

Abstract

We introduce HForge, a machine learning (ML) framework for predicting Hamiltonian (H) and Overlap (S) matrices directly from atomic structures, with a focus on amorphous (aBN) and hexagonal boron nitride (hBN). Leveraging graph-based descriptors derived from the MACE[1] model and reference Hamiltonians computed via Siesta, HForge enables efficient electronic structure predictions. In this poster we present how the choice of training structures impacts model performance and demonstrate that incorporating a diverse set of smaller structures significantly enhances the model's ability to generalize to larger systems—a key strategy given that training is 3–4 times more expensive than inference. We evaluate both equivariant and non-equivariant ML architectures, showing that equivariant models better preserve the physical symmetries of quantum interactions and outperform their non-equivariant counterparts in extrapolation tasks. Building on recent advancements in equivariant[2] graph-based atomic environment representations and universal message passing, our findings underscore the potential of scalable, ML-driven Hamiltonian prediction to accelerate classical DFT computations and enable quantum simulations of materials like aBN and hBN.

References

1. I. Batatia, D. P. Kovács, G. N. C. Simm, C. Ortner, and G. Csányi, arXiv preprint arXiv:2206.07697 (2023).
2. M. Geiger and T. Smidt, arXiv preprint arXiv:2207.09453 (2022).Authors, Journal, Issue (Year) page

Figures

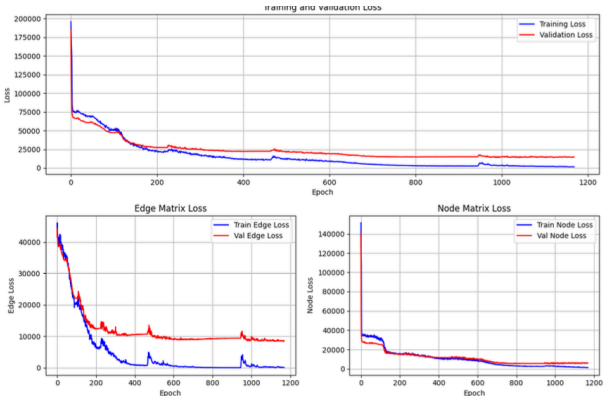

Figure 1: Mixed training on hBN and aBN structures containing 2, 3, 8, and 32 atoms shows that the model successfully fits the training set but still exhibits signs of overfitting. However, the performance improves dramatically compared to the sequential (independent) case shown in Figure 2.

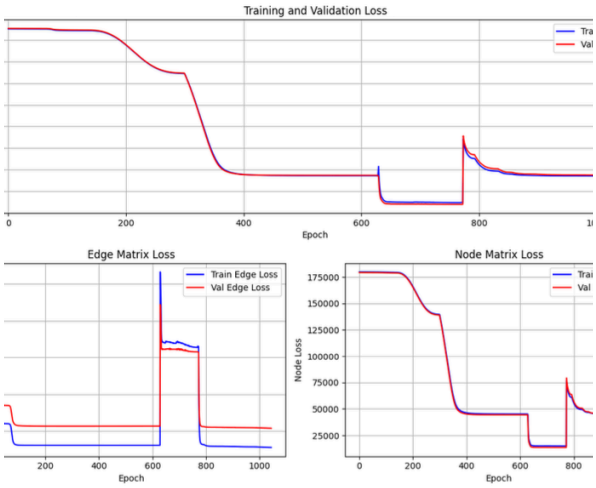

Figure 2: Sequential training with a fixed number of atoms: 32 → 3 → 32