Integrating Graph Neural Networks and Density Functional Theory in Many-Body Expansion Theory for High-Dimensional Potential Energy Surfaces

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In the present work we established an accurate and efficient computational tool that synergistically utilizes deep learning and quantum chemistry and predicts electronic structures and provides design principles for real-life functional materials like metal-organic frameworks (MOFs) and organic semiconductors (OSC). Density functional theory (DFT) has been a long-standing workhorse for evaluating electronic structures of these materials, but its computational complexity remains a great challenge for large systems with more than hundreds of atoms, even with significant approximation like many-body expansion (MBE). Here we integrated frontier graph neural network (GNN) models into the MBE theory to control the computational complexity and evaluated the high-dimensional ground- and excited-state potential energy surface for important functional materials. Technically, we broke down these materials into small repeating fragments, evaluated one-fragment energies using DFT but acquired two-fragment interactions and beyond based on GNN-trained structure–property relationships. The preliminary application of GNN-MBE on small organic molecules showed a remarkable reduction in the computational cost without compromising the accuracy and demonstrated a significant enhancement in both the performance and interpretability of GNN models in chemical systems. Leveraging the accuracy of DFT and the efficiency of GNN, the development of GNN-MBE marks a significant step towards computationally aided material design.