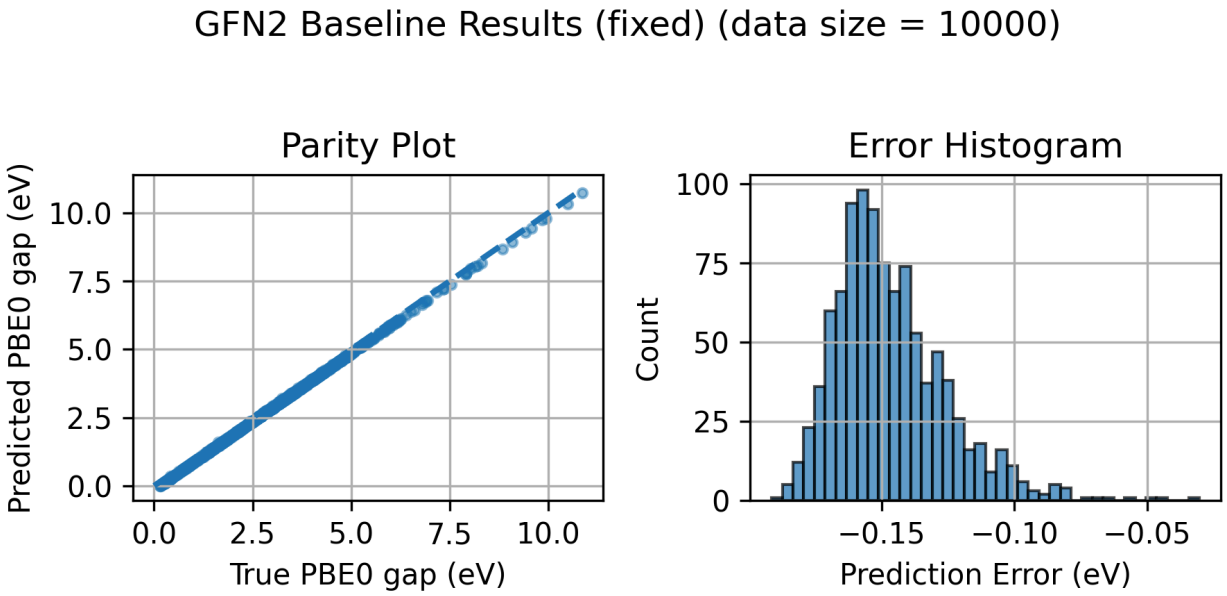
Predicting PBE0 HOMO–LUMO Gaps Using Graph Neural Networks and Δ-Learning

This report summarizes the results obtained from Task A (GFN2 baseline), Task B (Direct GNN), and Task C (Δ-GNN). The dataset consists of 10,000 molecules with computed GFN2 and PBE0 orbital gaps. Graph Neural Networks were trained to predict PBE0 either directly or using Δ-learning, where the model predicts the difference Δ = PBE0 - GFN2.

GFN2 Baseline Results (Task A)

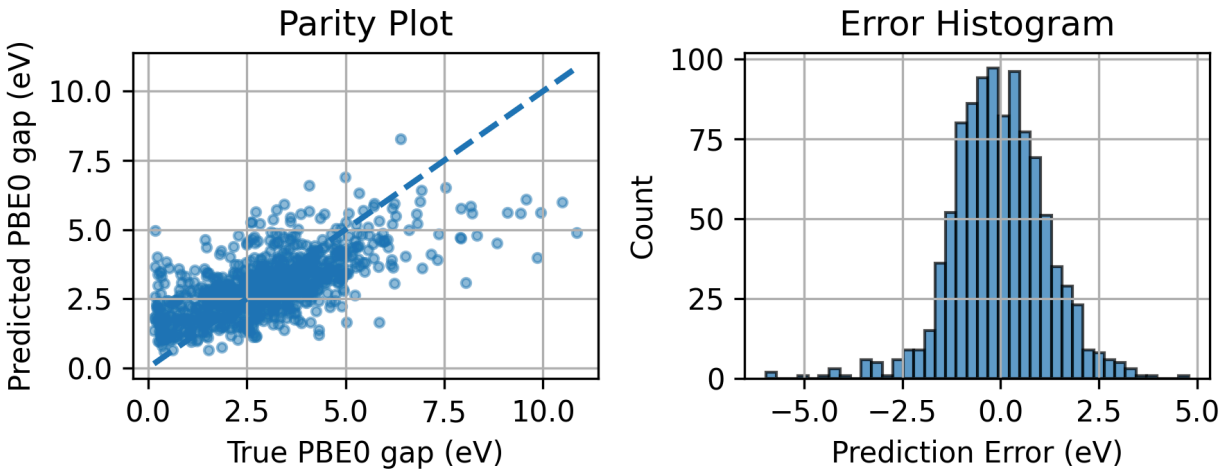
I



GFN2 exhibits linear behavior but with a clear negative bias, consistently underestimating the PBE0 gap. This systematic deviation highlights the need for machine learning correction such as Δ-learning.

Direct GNN Results (Task B)

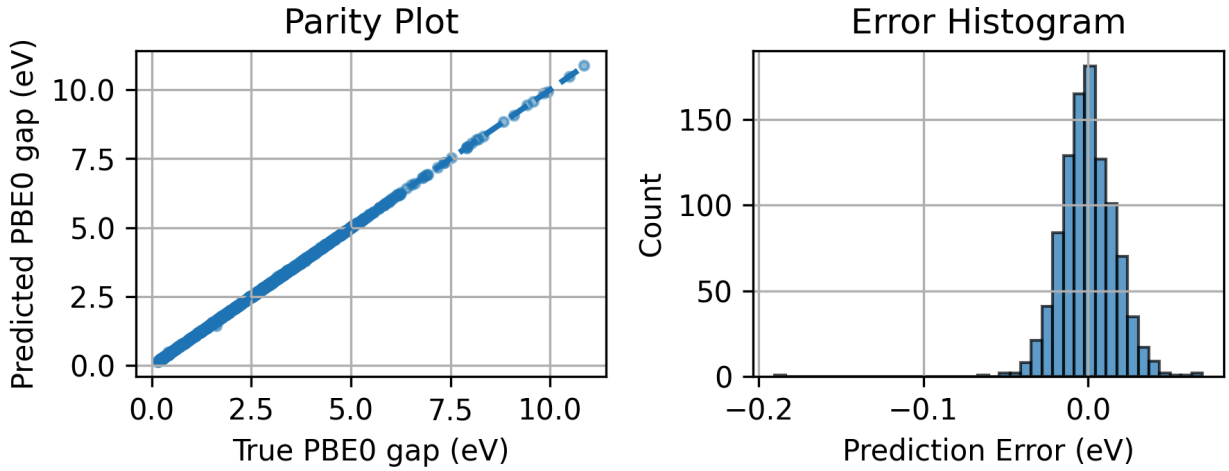
Direct GNN Results (fixed+extended) (data size = 10000)



Direct prediction of the PBE0 gap yields significant scatter and systematic underprediction. The model struggles with nonlinearity present in the full mapping from molecular graph to PBE0 gap. Validation errors remain relatively high.

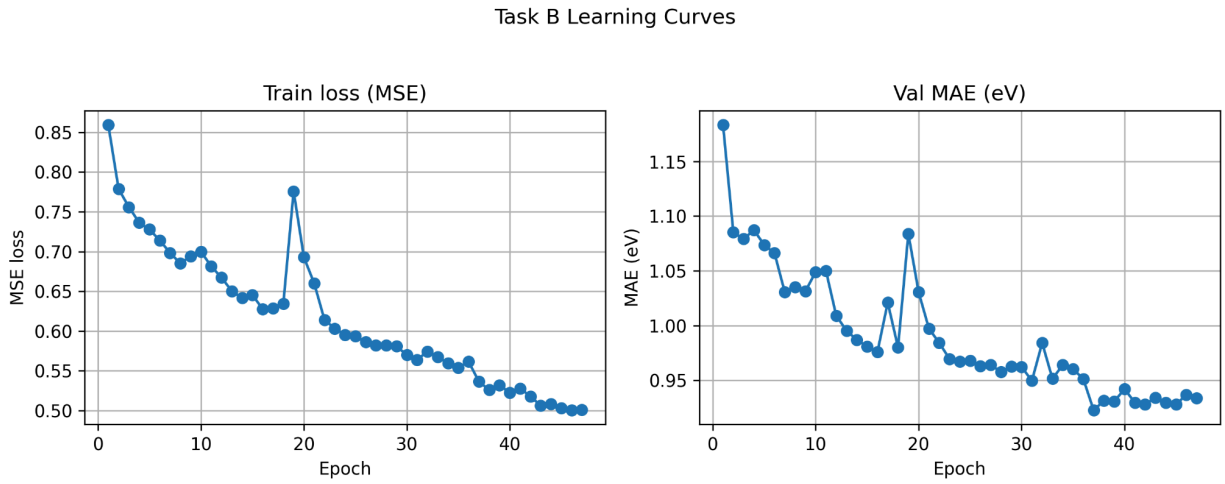
Δ-GNN Results (Task C)

Δ-GNN Results (fixed+extended) (data size = 10000)



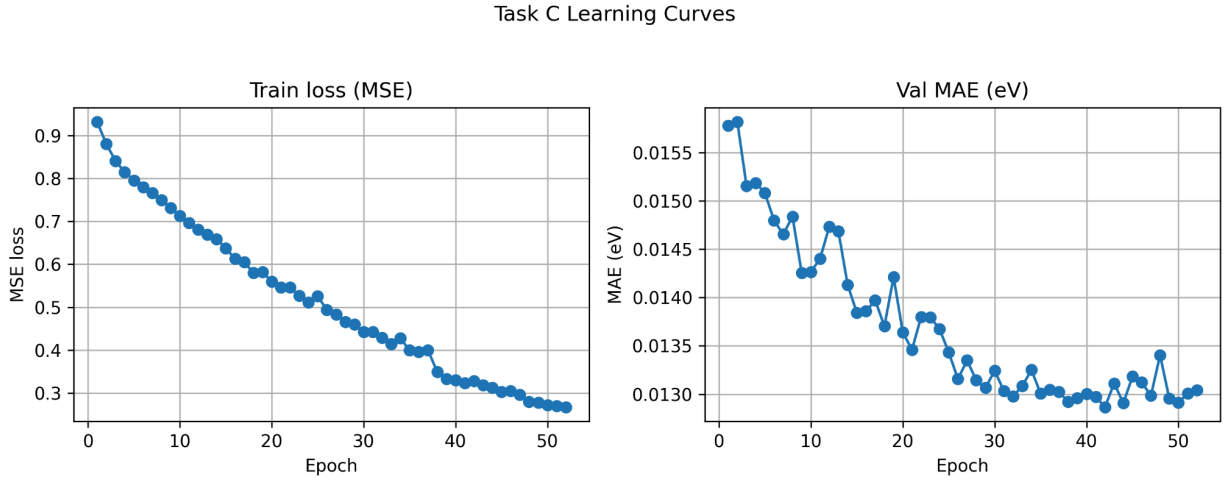
The Δ-GNN shows near-perfect predictions, aligning closely with the diagonal in the parity plot. The error histogram is sharply centered around zero, indicating excellent accuracy. This confirms that Δ-learning effectively captures the correction from GFN2 to PBE0.

Task B Learning Curves



Training loss decreases steadily but validation MAE fluctuates considerably, indicating unstable learning and difficulty predicting PBE0 directly from structure.

Task C Learning Curves



Δ-GNN exhibits smooth convergence in both training loss and validation MAE. The model quickly learns the residual correction function, achieving much lower error than the direct model.

Conclusion

From this experiment, we can clearly conclude that **Δ-learning is the most effective strategy for predicting PBE0 HOMO–LUMO gaps**, significantly outperforming both the GFN2 baseline and the direct GNN model. The GFN2 predictions, although roughly linear, show a strong systematic underestimation of PBE0 values. The direct GNN improves upon the raw baseline only slightly and exhibits large scatter and high error, especially for molecules with large gaps. This occurs because predicting the full PBE0 gap directly from structure is a highly nonlinear and complex task that requires the model to learn absolute electronic energies, long-range

quantum interactions, and subtle orbital physics purely from graph features—making the problem extremely difficult for a moderate-capacity GNN.

In contrast, the Δ-GNN model only needs to learn the *residual correction* between GFN2 and PBE0. This correction is smooth, low-magnitude, and chemically systematic (e.g., dependent on bond orders, atomic environments, and electronegativities). As a result, the Δ-GNN converges faster, achieves far lower error, and produces parity plots that lie almost perfectly on the diagonal. The learning curves further confirm that Δ-learning produces stable and monotonic convergence, whereas the direct model shows noisy and unstable optimization. Overall, the experiment demonstrates a key principle in computational chemistry ML: **learning the correction to a physically meaningful baseline (Δ-learning) is far easier and more accurate than learning an absolute quantum property from scratch**. This explains why Δ-learning has become the preferred method in modern molecular machine-learning pipelines.