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Towards a Transferable Acceleration Method for Density Functional Theory

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Motivation: The SCF Bottleneck

- **The Problem:** Density Functional Theory (DFT) relies on the iterative, costly Self-Consistent Field (SCF) method.
- Machine learning can provide a high-quality initial guess to reduce iterations.

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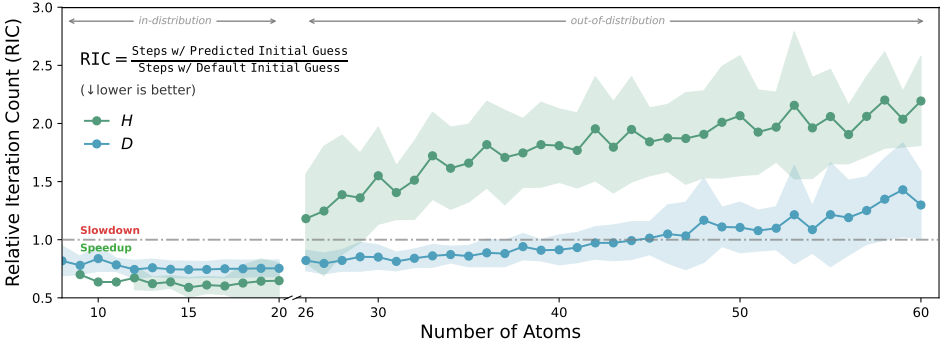
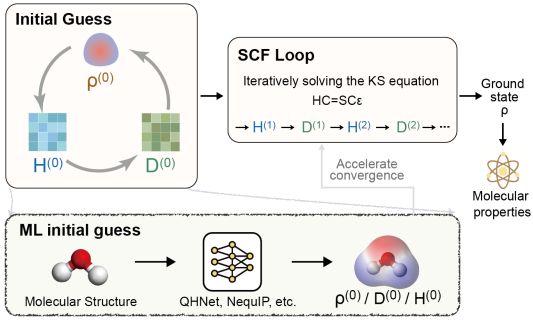
- **The Problem:** Density Functional Theory (DFT) relies on the iterative, costly Self-Consistent Field (SCF) method.
- Machine learning can provide a high-quality initial guess to reduce iterations.
- **Current Approaches:** Existing models target the Hamiltonian matrix, which is:
  - Numerically unstable to predict.
  - **Intrinsically non-transferable**: Fails on molecules larger than the training set.



Our Approach: Target the Electron Density

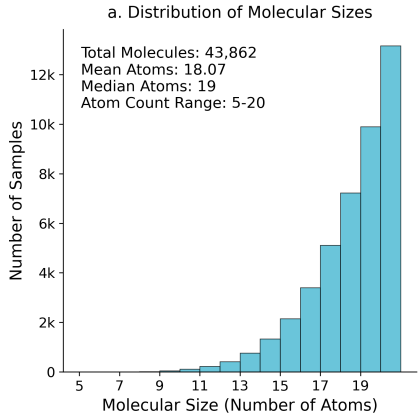
- **Core Insight:** The electron density (ρ) is a fundamental, local, and highly transferable property.
- **Methodology:**
  - Predict electron density in a *compact auxiliary basis* using E(3)-equivariant networks.
  - Use the prediction to assemble the initial Hamiltonian and Density matrix.
- **Advantage:** $O(N)$ scaling instead of $O(N^2)$, lower symmetry requirements, and data-efficient.



Introducing SCFbench

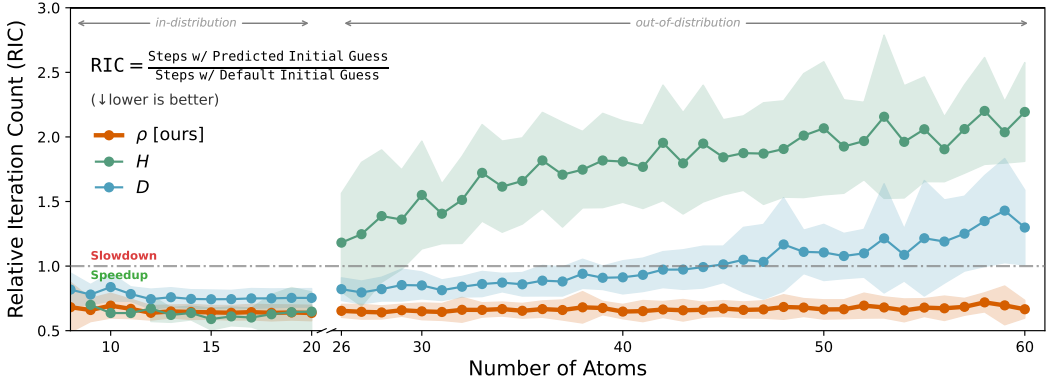
To facilitate research in transferable DFT acceleration, we introduce **SCFbench**:

- **Size:** 43,862 molecules (H, C, N, O, F, P, S).
- **What's Included:** Density matrices, Hamiltonians, and electron densities computed with multiple basis sets.
- **In-distribution Set:** Small molecules (≤ 20 atoms)
- **OOD Test Set:** Molecules up to 60 atoms to explicitly test size transferability.



Results: Zero-Shot Scalability

- **Robust Transferability:** Average **33.3% reduction** in SCF iterations on the out-of-distribution (60-atom) set.



- **Extreme Scaling:** Successfully accelerates massive systems *without retraining*:
  - Glycine-100: 703 atoms, converged in 10 iterations (17 for `minao`)
  - Polypropylene: 905 atoms, converged in 8 iterations (12 for `minao`)
- *Baselines ran out of memory or diverged at this scale.*

Conclusion

By shifting the prediction target from the Hamiltonian to the **electron density**, we unlock true size transferability for DFT acceleration.

- A single model trained on small molecules acts as a reliable "drop-in" accelerator for vast chemical spaces and various basis sets.
- **SCFbench** dataset and code are open-sourced.
  `https://huggingface.co/datasets/ByteDance-Seed/SCFbench`

Thank You!