Learning density functionals with differentiable DFT

Density functional theory (DFT) is a workhorse of quantum chemistry and materials science. Yet its predictive power remains limited by the quality of exchange-correlation (XC) functionals, which are traditionally designed through a combination of physical intuition, exact constraints and empirical fitting. Improving these density functional approximations is an active area of research in DFT.

Differentiable density functional theory offers a complementary method: instead of designing functionals by hand, it enables learning them directly from data while, in principle, retaining the structure of the underlying physics. By embedding machine learning models into the DFT framework and optimizing them end-to-end, this approach reframes functional development as a learning problem.

At its core, DFT involves solving the Kohn–Sham equations self-consistently to obtain an electron density and derived observables. The XC functional is the part of the Kohn–Sham density functional that captures the many-body electron interactions. In differentiable DFT, the XC functional is replaced by a learnable model, such as a neural network. Crucially, the entire self-consistent field (SCF) procedure is made differentiable. This means that gradients of the training loss with respect to model parameters can propagate through the SCF solver, without explicitly differentiating through each iteration. The functional is therefore not only evaluated within the Kohn–Sham framework but rather trained through it.

This end-to-end training through the Kohn–Sham equations enforces consistency with the governing equations at every step. As a result, differentiable DFT enables data-efficient learning and allows the construction of specialized ‘expert functionals’ tailored to particular classes of systems or properties. At the same time, physical constraints can be incorporated, for example through feature design, where model inputs are constructed to reflect known physics. This can further increase data efficiency, improve stability, and ensure physically consistent solutions.

These advantages come with challenges. Differentiating through the SCF procedure can be computationally demanding; if they are not carefully controlled, gradients may amplify instabilities in the self-consistency cycle. Regularization and robust solver implementations are therefore essential. Furthermore, although differentiable DFT promises improved transferability through the integrated physics, achieving this remains an active topic of research.

A central challenge in functional development is the treatment of non-locality. Whereas local and semi-local models, which depend only on the electron density at a point and its nearby variation, can be readily parametrized, it is considerably more difficult to capture long-range exchange-correlation effects in 3D. Traditionally, so-called hybrid functionals add a non-local exact exchange contribution computed directly from electron orbitals, making electron-electron interactions more explicit but increasing computational cost substantially. Recent advances in machine learning with graph-based models, attention mechanisms and operator-learning approaches, offer new

opportunities to develop non-local representations without sacrificing computational efficiency.

Differentiable DFT thus represents a shift in how density functionals can be developed and, in the broader sense, stands as an example of how to use differentiable programming to solve inverse problems. By turning the XC functional into a trainable model within a differentiable DFT framework, it enables learning quantum many-body effects directly from experimental or high-level quantum chemical data, while preserving known physics.

As methods and software progress, this approach may be used for systematic explorations of functional forms as well as for the development of explicitly non-local and application-specific functionals that were previously difficult to design by hand.

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