Orbital-free density functional theory for periodic solids: Construction of the Pauli potential

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The practical success of density functional theory (DFT) stems from the Kohn-Sham approach, which enables the exact calculation of the non-interacting electron kinetic energy via an auxiliary noninteracting system. Yet, the realization of DFT's full potential awaits the discovery of a direct link between the electron density, n and the non-interacting kinetic energy $T_S[n]$.

In this work, we address two key challenges towards this objective. First, we introduce a new algorithm for directly solving the constrained minimization problem to calculate $T_S[n]$ and its functional derivative at constant electron number, $V_S[n](\mathbf{r})$, for periodic densities. Second, we present a simple numerical procedure that allows us to calculate, for a given periodic density, the discontinuity in $V_S[n](\mathbf{r})$ which occurs upon increasing or decreasing the total number of electrons.

The code integrates key methodological innovations inspired by deep learning, such as implementation in JAX deep-learning framework, automatic differentiability, the use of an adaptive basis set ("equidensity orbitals") for wave function expansion and QR decomposition to accelerate the implementation of the orthogonality constraint. Notably, we derive a closed-form expression for the Pauli potential in one dimension, expressed solely in terms of the input density, without relying on Kohn-Sham eigenvalues and eigenfunctions. We validate this method on one-dimensional periodic densities, achieving results within chemical accuracy. We propose that our algorithm can be used to generate a database of energies, potentials, and densities, on which a neural network can be trained to arrive at an ultra-fast computation of the key functionals $T_S[n]$ and $V_S[n](\mathbf{r})$.