PWDFT.jl: A Julia package for electronic structure calculation using density functional theory and plane wave basis

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ABSTRACT

We describe the implementation of `PWDFT.jl`, a package for electronic structure calculations written using Julia programming language using plane wave basis set and pseudopotentials. In this package, typical Kohn-Sham density functional theory (KSDFT) is divided into three steps: instantiation of an atomistic models, initialization of Kohn-Sham Hamiltonian, and self-consistent field (SCF) calculations. `PWDFT.jl` is allows user to write their own SCF or alternative methods by providing implementation several basic operations such as wave function orthogonalization, action of kinetic and potential operators to wave functions and iterative diagonalization of Hamiltonian. Because `PWDFT.jl` is implemented in Julia, matrix operations within which plane wave KSDFT are formulated, are particularly easy to be implemented as used. With these features `PWDFT.jl` is expected to be useful for for beginners KSDFT implementers and researchers who want to implement their new methods quickly. `PWDFT.jl` is freely available at `https://github.com/f-fathurrahman/PWDFT.jl`.

Introduction

Electronic structure calculations based on Kohn-Sham density functional theory (KSDFT) [1, 2] is very popular nowadays. It is routinely used to model and predict various materials properties [3] by researchers from various fields such as physics, chemistry, and engineering. In practice, KSDFT calculations typically are carried out by using program packages such as Quantum Espresso [4], VASP [5], and ABINIT [6]. While using such program packages are very convenient for most practical purposes these packages arguably is not ideal for development. One factor which make development difficult for those program packages is due to the fact that they are written using static programming languages such Fortran and C/C++. Several attempts has been made to make development using dynamic programming languages such as Python (GPAW package [7]) and MATLAB (KSSOLV package [8]). However, the speed of execution of dynamic programming languages is generally slower than static programming language, so in order to obtain good performance one need to implement the time-consuming part of the code in C or Fortran which can hinder development of the code because programmers need to deal with low level details such as manual memory management.

Main Objectives

The main objective of this research is to implement a software package for doing Kohn-Sham density functional calculations with plane wave basis set and pseudopotentials. This software package should be simple yet able to carry out total energy calculations for non-trivial systems (not only toy models used in textbook).

Proposed solution

In this research we implement a software package named `PWDFT.jl` to carry out plane wave density functional calculations. This package is written in Julia programming language. Julia is a high-level dynamic programming language which has similarities to other dynamic languages such as MATLAB, R and Python.

As opposed to those dynamics language, Julia can achieve near-C-performance without having to write some portion of the code in C or Fortran. Julia also has first class support of multidimensional arrays like MATLAB which facilitates rapid development and quick prototyping for new algorithms.

Basic theory

In KSDFT approach [1, 2], total energy per unit cell system $E_{\text{total}}^{\text{KS}}$ can be written as

$$E_{\text{total}}^{\text{KS}} = E_{\text{kin}} + E_{\text{ele-nuc}} + E_{\text{Ha}} + E_{\text{xc}} + E_{\text{nuc-nuc}} \qquad (1)$$

with E_{kin} is the non-interacting kinetic energy of electrons, $E_{\text{ele-nuc}}$ is the interaction energy between electrons and nuclei, E_{Ha} is the classical electrostatic or Hartree energy, E_{xc} is the exchange-correlation energy, and $E_{\text{nuc-nuc}}$ is the interaction energy between nuclei. Central to the Kohn-Sham DFT is the one-electron (Kohn-Sham) orbitals $\psi_{i,\mathbf{k}}(\mathbf{r})$ with the associated orbital energies $\epsilon_{i,\mathbf{k}}$ and the electron density $\rho(\mathbf{r})$ which is obtained from the Kohn-Sham orbitals

$$\rho(\mathbf{r}) = \sum_{\mathbf{k}} \sum_{i=1}^{N_{\text{occ}}} w_{\mathbf{k}} f_{i,\mathbf{k}} \psi_{i,\mathbf{k}}^*(\mathbf{r}) \psi_{i,\mathbf{k}}(\mathbf{r}) \qquad (2)$$

$$\psi_{i,\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{\Omega}} \sum_{\mathbf{G}} c_{i,\mathbf{G}+\mathbf{k}} \exp\left[\mathrm{i}\left(\mathbf{G}+\mathbf{k}\right) \cdot \mathbf{r}\right] \qquad (3)$$

$$E_{\text{kin}} = -\frac{1}{2} \sum_{\mathbf{k}} \sum_i w_{\mathbf{k}} f_{i,\mathbf{k}} \int_{\Omega} \psi_{i,\mathbf{k}}^*(\mathbf{r}) \nabla^2 \psi_{i,\mathbf{k}}(\mathbf{r}) \,\mathrm{d}\mathbf{r} \qquad (4)$$

$$E_{\text{ele-nuc}} = \sum_{\mathbf{k}} \sum_i w_{\mathbf{k}} f_{i,\mathbf{k}} \int_{\Omega} \psi_{i,\mathbf{k}}^*(\mathbf{r}) V_{\text{ele-nuc}}(\mathbf{r}) \psi_{i,\mathbf{k}}(\mathbf{r}) \,\mathrm{d}\mathbf{r}. \qquad (5)$$

$$E_{\text{Ha}} = \frac{1}{2} \int_{\Omega} V_{\text{Ha}}(\mathbf{r}) \rho(\mathbf{r}) \,\mathrm{d}\mathbf{r} \qquad (6)$$

$$V_{\text{Ha}}(\mathbf{r}) = \int_{\Omega} \frac{\rho(\mathbf{r}')}{\mathbf{r}-\mathbf{r}'} \,\mathrm{d}\mathbf{r}' \qquad (7)$$

$$\nabla^2 V_{\text{Ha}}(\mathbf{r}) = -4\pi\rho(\mathbf{r}) \qquad (8)$$

$$E_{\text{xc}} = \int_{\Omega} \rho(\mathbf{r}) \epsilon_{\text{xc}}\left[\rho(\mathbf{r})\right] \,\mathrm{d}\mathbf{r} \qquad (9)$$

$$E_{\text{nuc-nuc}} = \frac{1}{2} \sum_{I \neq J} Z_I Z_J \left[\frac{\text{erfc}\left(\eta \left|\mathbf{R}_I - \mathbf{R}_J - \mathbf{T}\right|\right)}{\left|\mathbf{R}_I - \mathbf{R}_J - \mathbf{T}\right|} + \frac{4\pi}{\Omega} \sum_{\mathbf{G} \neq 0} \frac{1}{G^2} \exp\left(-\frac{G^2}{4\eta^2}\right) \cos\left(\mathbf{G} \cdot \left(\mathbf{R}_I - \mathbf{R}_J\right)\right) \right] \qquad (10)$$

Results

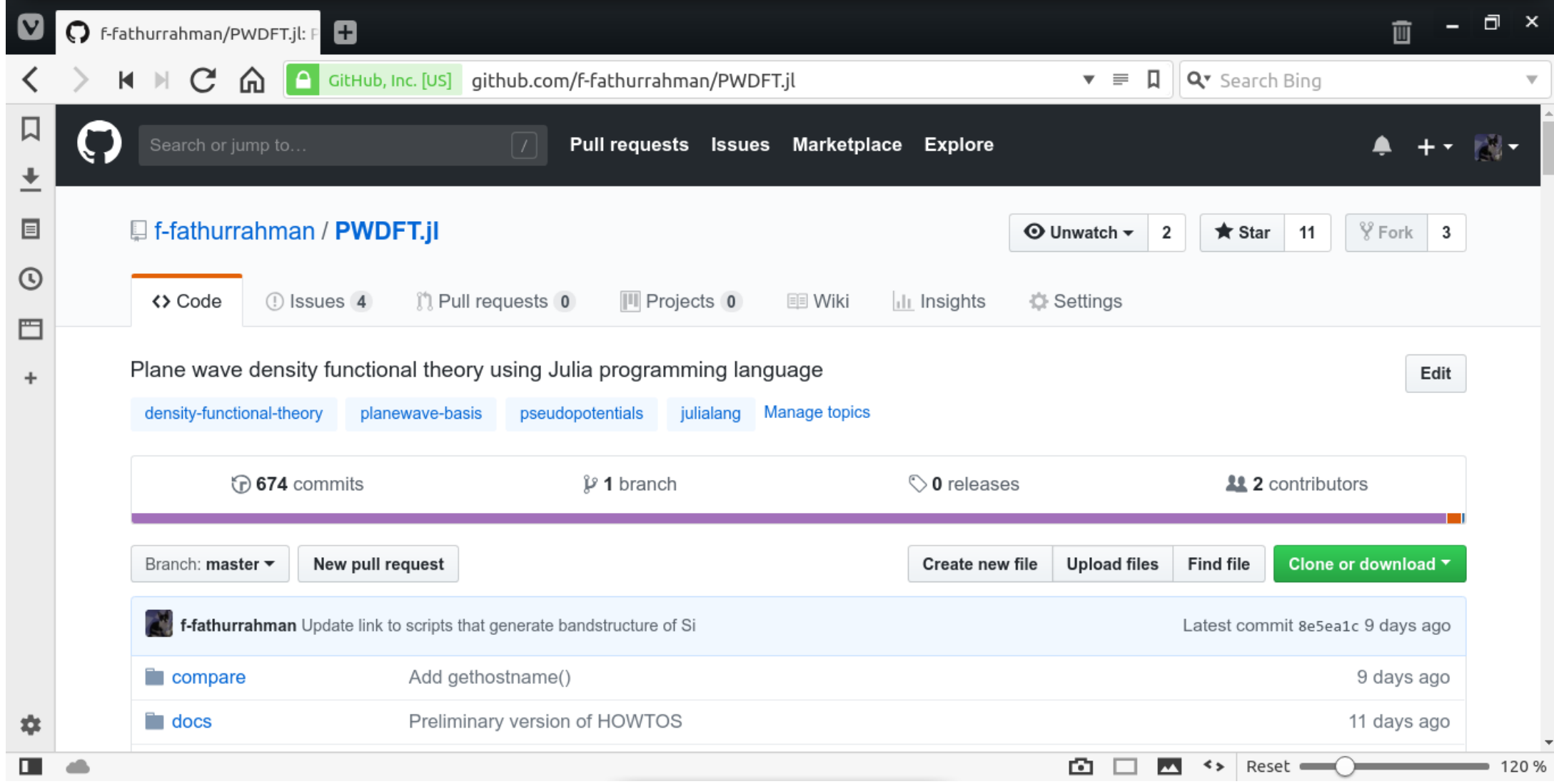The program package can be obtained from the Github repository at: `https://github.com/f-fathurrahman/PWDFT.jl`

The following code shows how one can calculate total energy of a molecule defined in an xyz file:

```julia
# Load the package
using PWDFT
# Stage 1: Initialization of atomic structures
atoms = Atoms(xyz_file="CH4.xyz")
# Stage 2: Initialization of Hamiltonian
ecutwfc = 15.0   # cutoff energy in hartree
pspfiles = ["C-q4.gth", "H-q1.gth"] # path to pseudopotential files
Ham = Hamiltonian( atoms, pspfiles, ecutwfc )
# Stage 3: Solve the Kohn-Sham problem
KS_solve_SCF!( Ham, betamix=0.2 )
```

Using `PWDFT.jl` one also can obtain various numerical data and processes it for visualization or other analysis. As an example, in Figure 2 highest occupied molecular orbitals of cobalt hydrocarbonyl HCo(CO)_4 is visualized.
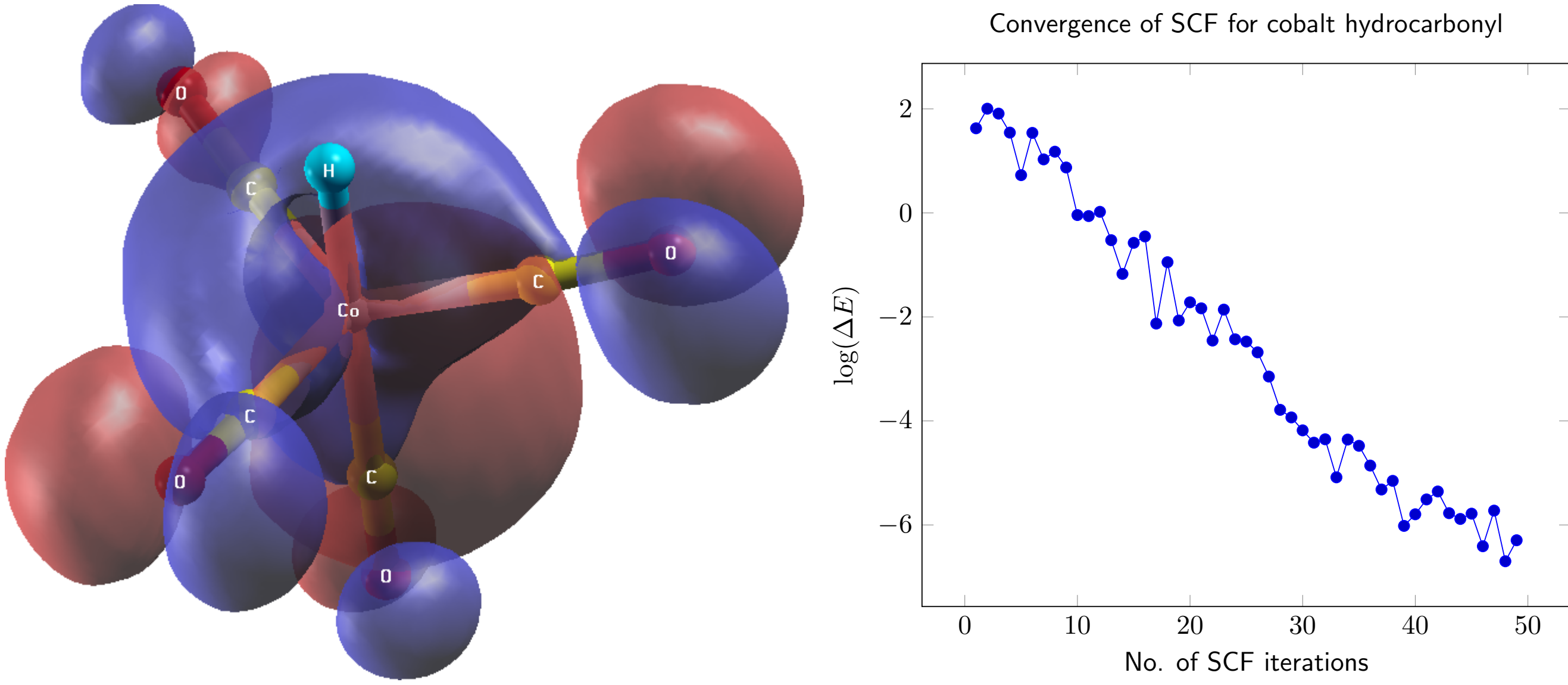

Figure 2: Highest molecular orbital of cobalt hydrocarbonyl (left) and progress of its SCF convergence (right)

Forthcoming Research

- Parallelization using MPI and CUDA
- Direct minimization algorithm for metallic systems

References

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