Introducing PROFESS 3.0: An advanced program for orbital-free density functional theory molecular dynamics simulations

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ABSTRACT

Orbital-free density functional theory (OFDFT) is a linear-scaling first-principles quantum mechanics method used to calculate the ground-state energy of a given system. Here we present a new version of PRinceton Orbital-Free Electronic Structure Software (PROFESS) with new features. First, PROFESS 3.0 provides a set of new kinetic energy density functionals (KEDFs) which are designed to model semiconductors or transition metals. Specifically, PROFESS 3.0 includes the Huang–Carter (HC) KEDF [1], a density decomposition method with fixed localized electronic density [2], the Wang–Govind–Carter (WGC) decomposition KEDF [3], and the Enhanced von Weizsäcker (EvW)-WGC KEDF [4]. Other major new functions are included, such as molecular dynamics with different statistical mechanical ensembles and spin-polarized density optimizers.

New version program summary

Program title: PROFESS

Catalogue identifier: AEBN_v3_0

Program summary URL: http://cpc.cs.qub.ac.uk/summaries/AEBN_v3_0.html

Program obtainable from: CPC Program Library, Queen's University, Belfast, N. Ireland

Licensing provisions: Standard CPC licence, http://cpc.cs.qub.ac.uk/licence/licence.html

No. of lines in distributed program, including test data, etc.: 76742

No. of bytes in distributed program, including test data, etc.: 1556428

Distribution format: tar.gz

Programming language: Fortran 90.

Computer: UNIX/Linux with ifort or gfortran.

Operating system: Linux.

Has the code been vectorized or parallelized?: Yes

RAM: Problem dependent, but 2 GB is sufficient for up to 10,000 ions using default settings.

Classification: 7.3.

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External routines: FFTW 3, Libxc 2.0.1., Lapack

Does the new version supersede the previous version?: Yes

Catalogue identifier of previous version: AEBN_v2_0

Journal reference of previous version: Comput. Phys. Comm. 181(2010)2208

Nature of problem:

Given a set of coordinates describing the initial ion positions under periodic boundary conditions, the problem is to determine the ground state energy, electron density, ion positions, and cell lattice vectors predicted by OFDFT. The computation of all terms is effectively linear scaling. Parallelization is implemented through domain decomposition, and up to around 10,000 ions may be included in the calculation on just a single processor, limited by RAM.

Solution method:

Two solutions based on computing energies as described in the text. The first method is to minimize the energy with respect to the electron density, ion positions, and cell lattice vectors. The second method is to perform molecular dynamics with different statistical mechanical ensembles.

Reasons for new version:

Conventional OFDFT performs excellently for light metals, such as aluminum, lithium, and magnesium. However, for semiconductors or transition metals, the required approximate non-interacting KEDFs and local pseudopotentials (LPSs) limit the accuracy of OFDFT. Here we present an updated version of PROFESS with a set of new KEDFs designed to model semiconductors or transition metals. Other newly developed functions, such as molecular dynamics and spin-polarized optimizers, are included.

Summary of revisions:

(a) Streamlining PROFESS: most module files in PROFESS 2.0 [5] are reconstructed, including splitting large modules into several small ones but with clear functions. These reconstructions improve the readability of PROFESS, reduce the compilation time, and make implementation and contribution of new functions to PROFESS easier for external developers.
(b) The format of the output file is improved to be more informative and clear.
(c) FFTW 2.1.5 is replaced with FFTW3-API interfaces, F95-API in serial mode, allowing for linking against either FFTW3 or a recent MKL and F2003-API in parallel mode.
(d) The spin polarized Perdew-Burke-Ernzerhof (PBE) [6] functional is implemented through the Libxc library [7].
(e) A subset of the latest Numerical Analysis Library (NMS) with improvements for F95 standard compliance is imported.
(f) The HC KEDF is fully supported (with force and stress evaluations) [1].
(g) A density decomposition method (DDM) with fixed localized electron density is supported (no force or stress yet) [2].
(h) The WGC [8] decomposition (WGCD) KEDF is supported (with force but no stress yet) [3].
(i) The EvW-WGC KEDF is supported (with force but no stress yet) [4].
(j) A new KEDF based on a point-wise Kohn–Sham [9, 10] kinetic energy density and electron localization function is supported (with force but no stress yet) [11].
(k) Spin-polarized calculations are supported [12]. Note that the reference density and therefore the kernels of the non-local KEDFs (except for the HC KEDF where the kernel for each spin channel is computed on-the-fly explicitly) are still simply derived from the total density, not from the electron density of each spin channel.
(l) The Limited-memory Broyden-Fletcher-Goldfarb-Shanno (L-BFGS) method [13, 14] is added for density optimization, which yields better stability and efficiency for spin-polarized and vacuum-containing systems.
(m) Molecular dynamics methods with three ensembles are implemented [15] and tested [16]: the microcanonical NVE (constant number of particles N, constant volume V, and constant energy E), the canonical NVT (constant number of particles N, constant volume V, and constant temperature T) with the Nosé-Hoover thermostat [17], and the isothermal–isobaric NPT (constant number of particles N, constant pressure P, and constant temperature T) with the Parrinello–Rahman thermostat [18].
(n) An option to set the initial density as a superposition of atomic densities is added.
(o) A bug in PROFESS 2.0 related to cutoff functions used to treat the vacuum region is fixed.
(p) The ion–ion interaction using the Ewald method (without the cubic b-spline method) is now parallelized.

Restrictions:

PROFESS cannot use non-local (such as ultrasoft) pseudopotentials. A variety of LPS files are available at the Carter group website (http://www.princeton.edu/carter/research/local-pseudopotentials).

Running time:

Problem dependent. Timing results for large-scale problems are given in the PROFESS paper [19].

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