Version 2.0.0 - M-SPARC: Matlab-Simulation Package for Ab-initio Real-space Calculations

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Abstract

M-SPARC is a MATLAB code for performing ab initio Kohn–Sham Density Functional Theory simulations. Version 2.0.0 of the software further extends its capability to include relativistic effects, dispersion interactions, and advanced semilocal/nonlocal exchange-correlation functionals. These features significantly increase the fidelity of first principles calculations that can be performed using M-SPARC.

Keywords: Kohn-Sham Density Functional Theory, Electronic structure, Relativistic effects, Dispersion interactions, meta-GGA functionals, Hybrid functionals

Metadata

\begin{tabular}{|c|c|}
\hline
C1 & Current code version \\
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& v2.0.0 \\
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C2 & Permanent link to code/repository used for this code version \\
\hline
& \url{https://github.com/SPARC-X/M-SPARC} \\
\hline
C3 & Code Ocean compute capsule \\
\hline
& N/A \\
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C4 & Legal Code License \\
\hline
& GNU General Public License v3.0 \\
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C5 & Code versioning system used \\
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& git \\
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C6 & Software code languages, tools, and services used \\
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& MATLAB 2013+ \\
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C7 & Compilation requirements, operating environments & dependencies \\
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& OS: Unix, Linux, MacOS, or Windows \\
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C8 & If available Link to developer documentation/manual \\
\hline
& \url{https://github.com/SPARC-X/M-SPARC/tree/master/doc} \\
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C9 & Support email for questions \\
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& phanish.s@gmail.com \\
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Refers to

Xu, Q., Sharma, A., and Suryanarayana, P., 2020. M-SPARC: MATLAB-Simulation Package for Ab-initio Real-space Calculations. SoftwareX, 11, 100423. \url{https://doi.org/10.1016/j.softx.2020.100423}

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Preprint submitted to arXiv December 15, 2022
1. Description of the software-update

M-SPARC [1] is an electronic structure code written in MATLAB that is based on real-space finite-difference method. It can perform spin-unpolarized and polarized ab initio calculations based on pseudopotential Kohn–Sham Density Functional Theory (DFT) [2, 3] for extended systems such as nanotubes/nanowires, surfaces, and crystals, as well as isolated systems such as molecules. In particular, the code can perform electronic ground state calculations for fixed atomic positions and cell dimensions (i.e., single-point calculations), geometry optimizations with respect to either atomic positions or cell volume, and microcanonical ensemble (NVE) molecular dynamics simulations, while employing norm-conserving pseudopotentials [4, 5]. In so doing, it can calculate the free energy, Hellmann–Feynman atomic forces, and Hellmann–Feynman stress tensor.

M-SPARC can be regarded as the MATLAB implementation of the large-scale parallel C/C++ code SPARC [6–8], with both codes employing the same algorithms, structure, input, and output. M-SPARC not only provides a suitable avenue for the first principles investigation of systems with small/moderate size, but also provides a prototyping platform that allows for the rapid development and testing of new methods/algorithms in real-space DFT [9–17], given the significant complexity of large-scale parallel codes written in lower level programming languages such as C/C++ and Fortran. Indeed, the development of KSSOLV [18] (MATLAB) and DFTK [19] (JULIA) for planewave DFT [20], and RESECU [21] (MATLAB) and RSDFT [22] (MATLAB) for real-space DFT have been similarly motivated.

Version 2.0.0 of the M-SPARC software further extends its capability to include relativistic effects, dispersion interactions, and advanced exchange-correlation functionals beyond the generalized gradient approximation (GGA) [20], as described below.

- **Spin-orbit coupling (SOC):** SOC is a relativistic effect that refers to the coupling between the electron’s orbital angular momentum and its spin angular momentum [20]. It becomes increasingly prominent for heavier atoms, i.e., those with larger atomic numbers, and is known to play a significant role in determining their electronic structure. M-SPARC incorporates SOC through relativistic norm-conserving pseudopotentials [23], as implemented within the real-space method [24].

- **Dispersion interactions:** Van der Waals (vdW) interaction is a correlation effect that arises due to the coupling in electronic charge fluctuations between different parts of the system [20]. This long-range dispersion interaction becomes increasingly important as the system gets more sparse, i.e., the inter-particle separation becomes larger. M-SPARC incorporates these interactions through the DFT-D3 correction [25] and the nonlocal vdW-density functional (vdW-DF) [26], as implemented using the method proposed in Ref. [27].

- **Meta-GGA functionals:** Meta-GGA exchange-correlation functionals represent the third rung of Jacob’s ladder, i.e., one rung above GGA [20]. Indeed, the sophistication and accuracy of the functionals increases as one goes up the ladder. In particular, in addition to the electron density and its gradient used to define GGA, the kinetic energy density is included in meta-GGA functionals. M-SPARC incorporates meta-GGA
through the SCAN functional \cite{SCAN}, which satisfies all seventeen constraints known on
the exact exchange-correlation functional.

- Hybrid exchange-correlation functionals: Hybrid exchange-correlation functionals rep-
  resent the fourth rung of Jacob’s ladder, i.e., one rung above meta-GGA, and therefore
two rungs above GGA \cite{GGA}. In particular, in addition to the semilocal GGA/meta-
GGA terms, a fraction of the Hartree-Fock exact exchange energy — quantity that
deeps explicitly on the occupied orbitals — is included in the exchange-correlation
functional. M-SPARC incorporates exact exchange through the PBE0 \cite{PBE0} and HSE
\cite{HSE} functionals, as implemented using the methods proposed in Refs. \cite{PBE0,HSE}.

In addition to the above, nonlinear core correction (NLCC) — accounts for the nonlinearity
in the exchange-correlation potential within pseudopotential generation — has been imple-
mented, a comprehensive testing framework with a large variety and number of examples has
been developed, and the table of SPMS \cite{SPMS} pseudopotentials — transferable and soft op-
timized norm-conserving Vanderbilt (ONCV) pseudopotentials \cite{ONCV} with NLCC for the PBE
\cite{PBE} variant of the GGA exchange-correlation functional — has been incorporated into the
M-SPARC distribution.

We now demonstrate the aforementioned major new functionalities of M-SPARC through
representative examples. Specifically, we consider (i) 2-atom primitive cell of body-centered
cubic (bcc) tantalum with PBE exchange-correlation, SOC through the relativistic ONCV
pseudopotential from the PseudoDOJO set \cite{PseudoDOJO}, \(6 \times 6 \times 6\) grid for Brillouin zone integration,
and mesh size of 0.14 Bohr; (ii) diazoxide molecule with PBE exchange-correlation, disper-
sion interactions through DFT-D3, and mesh-size of 0.24 Bohr; (iii) 14-atom cell of bulk
\(\text{Ni(CO}_2\text{)}_2\) with PBE exchange-correlation, dispersion interactions through vdW-DF, spin
polarization, \(2 \times 2 \times 2\) grid for Brillouin zone integration, and mesh size of 0.2 Bohr; (iv) 12-
atom (3,3) carbon nanotube with the SCAN variant of the meta-GGA exchange-correlation,
10 grid points for Brillouin zone integration, and a mesh size of 0.22 Bohr; and (v) 2-atom
primitive cell of germanene with HSE variant of hybrid exchange-correlation, \(4 \times 4\) grid for
Brillouin zone integration, and mesh size of 0.2 Bohr. Unless specified otherwise, we employ
ONCV pseudopotentials from the SPMS set. In all cases, we use 12-th order centered finite
differences for discretizing the equations, and perform single-point calculations. We present
the results so obtained in Fig. 1. To verify the accuracy of the results obtained by M-SPARC,
we compare them against highly converged results obtained using the established planewave
codes ABINIT \cite{ABINIT} and Quantum Espresso (QE) \cite{QE}. It is clear that there is excellent agree-
ment between M-SPARC and ABINIT/QE, verifying the accuracy of the M-SPARC code.
Indeed, the agreement further increases on refining the discretization, i.e., choosing smaller
values for the mesh-size.

The new functionalities in M-SPARC v2.0.0 allow for first principles simulations with
significantly higher fidelity compared to v1.0.0. Given that some of these features, in partic-
ular those involving meta-GGA and hybrid functionals, are noticeably more expensive than
standard GGA, new methods/algorithms to accelerate such calculations are highly desired,
for which M-SPARC provides a convenient avenue for rapid prototyping.
Figure 1: Examples demonstrating the major new functionalities of M-SPARC v2.0.0.
Acknowledgements

This work was supported by grant DE-SC0019410 funded by the U.S. Department of Energy, Office of Science. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the Department of Energy, or the U.S. Government.

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