

Assessing the source of error in local orbital-free density functionals

Bishal Thapa,^{1,2} Xin Jing,^{3,4} John E. Pask,⁵ Phanish Suryanarayana,³ and Igor I. Mazin^{1,2, a)}

¹⁾*Department of Physics and Astronomy, George Mason University, Fairfax, VA 22030, USA*

²⁾*Quantum Science and Engineering Center, George Mason University, Fairfax, VA 22030, USA*

³⁾*College of Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA*

⁴⁾*College of Computing, Georgia Institute of Technology, Atlanta, GA 30332, USA*

⁵⁾*Physics Division, Lawrence Livermore National Laboratory, Livermore, California 94550, USA*

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We investigate the source of error in local orbital-free density functionals relative to Kohn-Sham density functional theory (DFT). In particular, through numerical studies on a range of materials, for a variety of crystal structures subject to strain and atomic displacements, we find that while the ground state electron density in Thomas-Fermi-von Weizsäcker (TFW) orbital-free DFT is close to the Kohn-Sham density, the corresponding energy deviates significantly from the Kohn-Sham value. We show that these differences are a consequence of the poor representation of the linear response susceptibility within the TFW approximation for the electronic kinetic energy, confirming conjectures in the literature. In so doing, we find that the energy computed from a non-self-consistent Kohn-Sham calculation using the TFW electronic ground state density is in very good agreement with that obtained from the fully self-consistent Kohn-Sham solution.

I. INTRODUCTION

Density functional theory (DFT)^{1,2} is one of the most widely used ab initio methods in physical, chemical, and materials science research for understanding and predicting the properties of materials systems. Its conceptual foundation lies in the Hohenberg-Kohn (HK) theorem³, which states that the total energy of the system is a unique, albeit unknown, functional of its density. This rather formal mathematical concept was made practically useful by the Kohn-Sham (KS) formalism⁴, wherein the real system of interacting electrons is replaced by a fictitious system of non-interacting fermions that generates the same electronic density. In particular, the electronic kinetic energy is no longer an explicit functional of the density, but rather takes the form (in atomic units):

$$T_s = -\frac{1}{2} \sum_{n=1}^{N_s} \int \psi_n(\mathbf{r}) \nabla^2 \psi_n(\mathbf{r}) d\mathbf{r},$$

where $\{\psi_n\}_{n=1}^{N_s}$ are the KS orbitals. In so doing, a one-electron Schrödinger-type equation needs to be solved for multiple electronic states, i.e., Kohn-Sham orbitals, whose number grows with the system size, which together with the orthogonality constraint on the orbitals, results in computations that scale cubically with system size¹. This severely restricts the range of systems that can be studied using KS-DFT.

An alternative to replacing the system of interacting electrons with a fictitious system of non-interacting

fermions is to replace it instead with a fictitious system of non-interacting bosons. This can be achieved by approximating the kinetic energy T_s using an explicit functional of the density, the resulting formalism referred to as orbital-free (OF) DFT⁵. This generally amounts to solving a Schrödinger-type equation for only one electronic state, which corresponds to the square root of the density. Though the computational cost of OF-DFT scales linearly with system size, thereby overcoming the cubic-scaling bottleneck of KS-DFT, it is rarely used in practice due to the lack of accurate approximations for the kinetic energy T_s .

Historically, the first approximation for T_s was suggested long before the advent of DFT, by Thomas⁶ and Fermi⁷:

$$T_{TF} = \int \left(\frac{3}{10} (3\pi^2)^{\frac{2}{3}} \rho(\mathbf{r})^{\frac{5}{3}} \right) d\mathbf{r},$$

which is exact in the limit of slowly varying density. The first gradient correction was derived by von Weizsäcker⁸:

$$T_{\lambda W} = \int \left(\frac{\lambda}{8} \frac{|\nabla \rho(\mathbf{r})|^2}{\rho(\mathbf{r})} \right) d\mathbf{r},$$

the weight factor being $\lambda = 1$, which represents a lower bound on the kinetic energy. This term can be shown to be exact in the limit of small and rapid (large wavevector) density variations⁹. In the opposite limit of slow (but not necessarily small) variations, a similar formula can be derived¹⁰, with weight factor instead being $\lambda = 1/9$. There have been suggestions to use other values for the weight factor² as well as a position dependent weight factor¹¹, however such strategies depend upon the class of system under consideration and so lack universality.

^{a)}Email: imazin2@gmu.edu

The dramatic difference between the limiting values for the weight factor in $T_{\lambda W}$ signals the inability of local functionals of the density and its gradient to describe the kinetic energy T_s even for small density variations, if the scale of variations is neither particularly large nor particularly small. This led to thinking, as early as three decades ago¹², of the need for functionals that are nonlocal in coordinate space, i.e., depend on the density correlation at finite distances, e.g.:

$$T_s = T_{TF} + \int \rho(\mathbf{r})K(\rho(\mathbf{r}), \rho(\mathbf{r}'))\rho(\mathbf{r}') d\mathbf{r} d\mathbf{r}',$$

where the nonlocal kernel $K(\rho(\mathbf{r}), \rho(\mathbf{r}'))$, is selected in such a way so as to exactly reproduce the inverse electronic susceptibility — given by the Lindhard formula¹³, $\chi_{Lind}(\mathbf{r}, \mathbf{r}') = 1/\chi(\mathbf{r}, \mathbf{r}') = \delta^2 E_{KS}/\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')$ — in the uniform electron gas limit. Note that a slightly different form essentially equivalent to that presented above has also been proposed¹²: $T_s = T_{TF} + T_W[\tilde{\rho}(\mathbf{r})]$, where $\tilde{\rho}(\mathbf{r}) = \int \rho(\mathbf{r}')K(\rho(\mathbf{r}), \rho(\mathbf{r}')) d\mathbf{r}'$. In subsequent work¹⁴, it was pointed out that a form more consistent with the idea of generalizing the von Weizsäcker-Kirzhnits term into the domain of nonlocal functionals should include density log-gradients in powers adding up to 2, e.g.,

$$T_s = T_{TF} + \int \frac{\nabla\rho(\mathbf{r})}{\rho(\mathbf{r})}K(\rho(\mathbf{r}), \rho(\mathbf{r}'))\frac{\nabla\rho(\mathbf{r}')}{\rho(\mathbf{r}')}d\mathbf{r} d\mathbf{r}'.$$

A similar idea was later proposed by Wang and Teter¹⁵, who represented the kinetic energy as

$$T_s = T_{TF} + T_{\lambda W} + \int \rho(\mathbf{r})^a K(\mathbf{r}, \mathbf{r}')\rho(\mathbf{r}')^b d\mathbf{r} d\mathbf{r}',$$

where $a + b = 8/3$ ensures the correct dimensionality. This was later generalized to density dependent kernels by Wang, Govind, and Carter (WGC)¹⁶:

$$T_s = T_{TF} + T_{\lambda W} + \int \rho(\mathbf{r})^a K(\rho(\mathbf{r}), \rho(\mathbf{r}'))\rho(\mathbf{r}')^b d\mathbf{r} d\mathbf{r}'.$$

While showing good results for particular problems^{16–23}, such nonlocal functionals have found rather limited use in practice due to greater computational expense and the need for specialized kernels to be developed for different materials systems^{18,20–22,24}. Further advances require a more fundamental understanding of linear response, and in particular, whether it is the deciding factor in determining the error associated with OF-DFT, as conjectured in previous works^{12,14,15,25}.

The orbital-corrected orbital-free (OO) DFT approach was recently proposed²⁶, wherein the ground state electron density obtained from a WGC OF-DFT calculation is used as input for a single self-consistent field (SCF) iteration of a KS calculation, the quantities so obtained then being used to compute the energy. This can be understood as performing a non-self-consistent KS calculation with the electronic ground state density

from WGC OF-DFT as input. It was found that the OO-DFT ground state energy is in very good agreement with that from KS-DFT. This interesting proposal was, however, largely empirical, in contrast to the above-mentioned nonlocal functionals underpinned by linear response theory. Furthermore, WGC already incorporates some amount of linear response, complicating the ability to draw clear inferences. Finally, the empirical evidence consists of just two cases, namely the energy-volume curves for fcc Ag and cubic-diamond Si. To what extent this idea is universal, and how (if at all) it is related to linear response theory, remain to be clarified.

In this paper, we address the aforementioned issues. First, we verify that the “single-shot” KS calculation with TFW OF-DFT ground state density as input is close to the fully self-consistent KS-DFT solution for a range of materials, including different crystal structures subject to volumetric and symmetry-lowering perturbations. Second, we argue that the success of such a strategy indicates that the main limitation of OF-DFT is indeed its inability to properly describe the correct linear response, as conjectured in the literature^{12,14,15,25}.

II. SYSTEMS AND METHODS

We consider body-centered cubic (BCC), face-centered cubic (FCC), hexagonal close packed (HCP), and body-centered tetragonal (BCT) crystals of magnesium (Mg), aluminum (Al), and indium (In); as well as diamond cubic (DC) and hexagonal diamond (DH) (also known as lonsdaleite) crystals of silicon (Si). These systems form a diverse set that includes a simple metal, transition metal, and semiconductor, in a variety of lattice configurations. Importantly, well-tested local pseudopotentials are available for the chemical elements in question, i.e., Mg, Al, In, and Si, allowing for a careful comparison of the results obtained from KS-DFT and OF-DFT calculations.

Unless specified otherwise, we choose the primitive unit cells for each of the systems, i.e., 1-atom cells for the BCC and FCC lattices, 2-atom cells for the HCP, BCT, and DC lattices, and 4-atom cells for the DH lattice. For the HCP and BCT lattices, we choose the ideal c/a ratios 1.633 and 1.414, respectively. After determining the equilibrium configurations, we consider the following strains and atomic displacements:

- Volumetric strains for each of the aforementioned systems. In this case, the strain tensor takes the form:

$$\mathbf{G} = \begin{pmatrix} 1+g & 0 & 0 \\ 0 & 1+g & 0 \\ 0 & 0 & 1+g \end{pmatrix}, \quad (1)$$

where the perturbation parameter $-0.1 \leq g \leq 0.1$, with $g < 0$ and $g > 0$ corresponding to the contraction and expansion of the unit cell, respectively.

- Symmetry-lowering, volume-preserving rhombohe-

dral strains for Mg, Al, and In in the FCC and BCC crystal configurations; and Si in the DC crystal configuration. In this case, the strain tensor takes the form:

$$\mathbf{G} = (1 + 3g)^{-\frac{1}{3}} \begin{pmatrix} 1 + g & g & g \\ g & 1 + g & g \\ g & g & 1 + g \end{pmatrix}, \quad (2)$$

where the perturbation parameter $-0.1 \leq g \leq 0.1$, with $g < 0$ and $g > 0$ corresponding to the compression and elongation of the unit cell, respectively, along the [111] direction.

- Volume-preserving uniaxial strains along the [001] direction for Mg, Al, and In in the HCP and BCT crystal configurations; and Si in the DH crystal configuration. In this case, the strain tensor takes the form:

$$\mathbf{G} = \begin{pmatrix} (1 + g)^{-\frac{1}{2}} & 0 & 0 \\ 0 & (1 + g)^{-\frac{1}{2}} & 0 \\ 0 & 0 & 1 + g \end{pmatrix}, \quad (3)$$

where the perturbation parameter $-0.1 \leq g \leq 0.1$, with $g < 0$ and $g > 0$ corresponding to the compression and elongation of the unit cell, respectively.

- Symmetry-lowering atomic perturbations (i.e., frozen phonons) for Al and Mg in the BCC, FCC, and HCP crystal configurations; and In in the FCC, HCP, and BCT crystal configurations. For BCC and FCC, we choose the 2-atom conventional cell and 2-atom tetragonal cells, respectively, rather than the 1-atom primitive cell used in other simulations. In all cases, the atom is perturbed along the [001] direction, with the z coordinate of one of the atoms changed as: $z \rightarrow (1 + g)z$, where the perturbation parameter $-0.05 \leq g \leq 0.05$.

All calculations are performed using the M-SPARC code^{27,28}, which is a `Matlab` version of the large-scale parallel electronic structure code, SPARC²⁹. It employs the real-space finite-difference method, whose formulation and implementation in the context of KS-DFT and OF-DFT can be found in previous works^{30–33}. We employ the local density approximation (LDA)^{4,34} for the exchange-correlation functional and use the bulk-derived local pseudopotentials (BLPS)³⁵. In the OF-DFT calculations, we choose the TFW kinetic energy functional with weight factor $\lambda = 1/8$. In the KS-DFT calculations, we perform Brillouin zone integration using a $15 \times 15 \times 15$ Monkhorst-Pack grid for the FCC, BCC, DC, and DH lattices, and $15 \times 15 \times 10$ grid for the HCP and BCT lattices, which ensures that the energies are converged to within 10^{-4} ha/atom. In all calculations, we employ a 12-th order finite-difference approximation and a grid spacing of 0.4 bohr, which ensures that the computed energies are converged to within 10^{-3} ha/atom. Finally, the change in energy arising due to a perturbation, which

is the main quantity of interest in the present work (Section III), is converged to within 10^{-6} ha/atom.

III. RESULTS AND DISCUSSION

We use the framework described in the previous section to perform KS-DFT and OF-DFT calculations for the selected systems. In particular, for each system, we compute the four energies listed below.

- $E_{KS \rightarrow KS} := E_{KS}(\rho_{KS})$: KS-DFT energy E_{KS} corresponding to the KS-DFT ground state density ρ_{KS} . This is obtained by performing a standard electronic ground state calculation in KS-DFT.
- $E_{OF \rightarrow KS} := E_{KS}(\rho_{OF})$: KS-DFT energy E_{KS} corresponding to the OF-DFT ground state density ρ_{OF} . This involves the calculation of orbitals for the given ρ_{OF} , i.e., a single self-consistent field (SCF) iteration in KS-DFT.
- $E_{KS \rightarrow OF} := E_{OF}(\rho_{KS})$: OF-DFT energy E_{OF} corresponding to the KS-DFT ground state density ρ_{KS} .
- $E_{OF \rightarrow OF} := E_{OF}(\rho_{OF})$: OF-DFT energy E_{OF} corresponding to the OF-DFT ground state density ρ_{OF} . This is obtained by performing a standard electronic ground state calculation in OF-DFT.

To quantify the error in the energies $E_{OF \rightarrow KS}$, $E_{KS \rightarrow OF}$, and $E_{OF \rightarrow OF}$, the error being defined with respect to $E_{KS \rightarrow KS}$, we define the following root-mean-square measure:

$$\Delta = \sqrt{\frac{\int (\Delta E(g) - \Delta E_{KS \rightarrow KS}(g))^2 dg}{\int dg}}, \quad (4)$$

where

$$\begin{aligned} \Delta E(g) &= E(g) - E(0), \\ \Delta E_{KS \rightarrow KS}(g) &= E_{KS \rightarrow KS}(g) - E_{KS \rightarrow KS}(0), \end{aligned}$$

with $E \in \{E_{OF \rightarrow KS}, E_{KS \rightarrow OF}, E_{OF \rightarrow OF}\}$, and the corresponding $\Delta \in \{\Delta_{OF \rightarrow KS}, \Delta_{KS \rightarrow OF}, \Delta_{OF \rightarrow OF}\}$. Note that the difference in energies from $g = 0$ is used in the definition of the error since the reference energy within KS-DFT and OF-DFT is different, a consequence of the different energy functionals, i.e., only differences in energy within the same level of theory are meaningful.

In Fig. 1 and Table I, we summarize the results so obtained, with the detailed data available in the Supplementary Material. We observe the following trend in the Δ -errors: $\Delta_{OF \rightarrow KS} < \Delta_{KS \rightarrow OF} < \Delta_{OF \rightarrow OF}$. In particular, the values of $\Delta_{OF \rightarrow KS}$ are quite small, which suggests that the ground state density in OF-DFT is close to that in KS-DFT. Furthermore, since the values of $\Delta_{KS \rightarrow OF}$ are relatively large, it can be inferred that the energy errors in OF-DFT are not a consequence of the errors in the ground state density, but are rather due to a fundamental limitation in the energy functional.

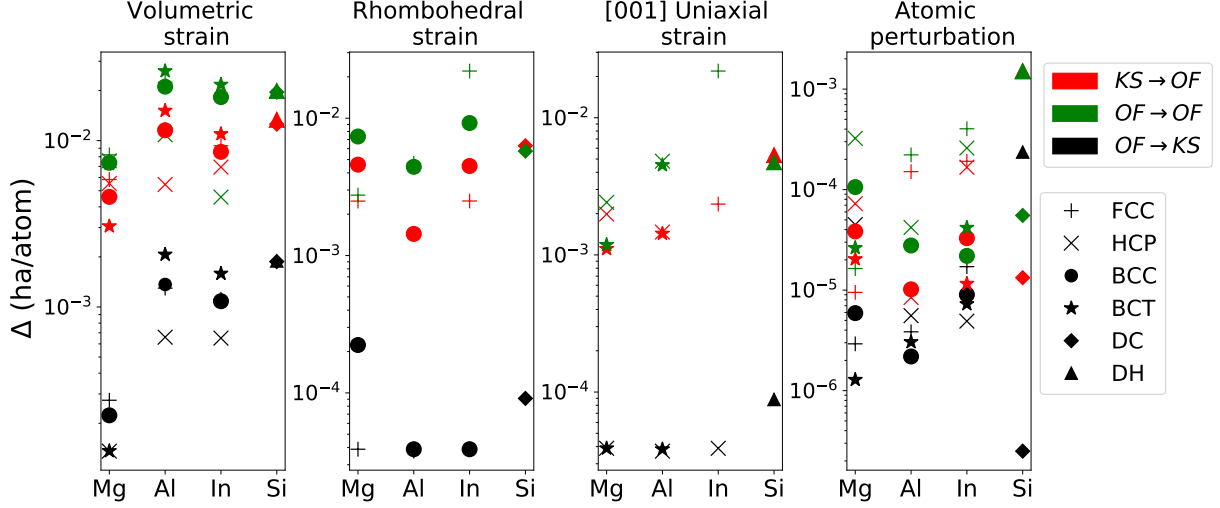


FIG. 1: Δ -error in the $E_{OF \rightarrow KS}$, $E_{KS \rightarrow OF}$, and $E_{OF \rightarrow OF}$ energies.

The above findings can be understood in terms of linear response theory:

$$\begin{aligned} \Delta E_{KS \rightarrow KS}(g) &= E_{KS}(\rho_{KS}(g)) - E_{KS}(\rho_{KS}(0)) \\ &= \int \chi_{KS}(\mathbf{r}, \mathbf{r}') \Delta \rho_{KS}(\mathbf{r}) \Delta \rho_{KS}(\mathbf{r}') d\mathbf{r} d\mathbf{r}' \end{aligned} \quad (5)$$

$$\begin{aligned} \Delta E_{OF \rightarrow KS}(g) &= E_{KS}(\rho_{OF}(g)) - E_{KS}(\rho_{OF}(0)) \\ &= \int \chi_{KS}(\mathbf{r}, \mathbf{r}') \Delta \rho_{OF}(\mathbf{r}) \Delta \rho_{OF}(\mathbf{r}') d\mathbf{r} d\mathbf{r}' \end{aligned} \quad (6)$$

$$\begin{aligned} \Delta E_{KS \rightarrow OF}(g) &= E_{OF}(\rho_{KS}(g)) - E_{OF}(\rho_{KS}(0)) \\ &= \int \chi_{OF}(\mathbf{r}, \mathbf{r}') \Delta \rho_{KS}(\mathbf{r}) \Delta \rho_{KS}(\mathbf{r}') d\mathbf{r} d\mathbf{r}' \end{aligned} \quad (7)$$

$$\begin{aligned} \Delta E_{OF \rightarrow OF}(g) &= E_{OF}(\rho_{OF}(g)) - E_{OF}(\rho_{OF}(0)) \\ &= \int \chi_{OF}(\mathbf{r}, \mathbf{r}') \Delta \rho_{OF}(\mathbf{r}) \Delta \rho_{OF}(\mathbf{r}') d\mathbf{r} d\mathbf{r}' \end{aligned} \quad (8)$$

where

$$\Delta \rho_{KS} = \rho_{KS}(g) - \rho_{KS}(0), \quad (9)$$

$$\Delta \rho_{OF} = \rho_{OF}(g) - \rho_{OF}(0), \quad (10)$$

with χ_{KS} and χ_{OF} being the linear response susceptibilities in KS-DFT and OF-DFT, respectively. Since the difference between $\Delta E_{KS \rightarrow KS}$ and $\Delta E_{OF \rightarrow KS}$ is small, as found in the numerical results above, it follows from Eqs. 5 and 6 that the ground state densities of KS-DFT and OF-DFT are *close*, which also justifies the use of linear response theory. Furthermore, since the difference between $\Delta E_{KS \rightarrow OF}$ and $\Delta E_{KS \rightarrow KS}$ is relatively large, as found in the numerical results above, it follows from Eqs. 5 and 7 that the error in the energy for OF-DFT calculations is due to the poor representation of the linear response susceptibility χ_{OF} relative to χ_{KS} , which motivates the need for developing alternate functionals with better linear response.

	$\Delta_{OF \rightarrow KS}$	$\Delta_{KS \rightarrow OF}$	$\Delta_{OF \rightarrow OF}$
Volumetric strain	0.001025	0.008895	0.015246
Rhombohedral strain	0.000046	0.003087	0.007532
[001] Uniaxial Strain	0.000045	0.002511	0.006758
Atomic perturbation	0.000026	0.000171	0.000235

TABLE I: Average Δ -error in the $E_{OF \rightarrow KS}$, $E_{KS \rightarrow OF}$, and $E_{OF \rightarrow OF}$ energies..

The above results also suggest a possible strategy to accelerate KS-DFT calculations without significant loss of accuracy. In particular, the ground state density computed from OF-DFT, which scales linearly with system size, can be used as input to perform a single SCF iteration in KS-DFT or to accelerate SCF convergence. Indeed, for such a strategy to be generally applicable, nonlocal pseudopotentials, which are the standard in KS-DFT, need to be incorporated into OF-DFT²³.

IV. CONCLUDING REMARKS

In this work, we have systematically investigated the source of error arising in local orbital-free density functionals relative to Kohn-Sham DFT. In particular, through numerical studies on a variety of materials, for a range of crystal structures subject to strains and atomic displacements, we have found that while the ground state electron density in the TFW variant of orbital-free DFT is close to the Kohn-Sham ground state density, the corresponding energy differs significantly from the Kohn-Sham value. We have shown that these differences arise due to the poor representation of the linear response susceptibility within the TFW approximation for the electronic kinetic energy, therefore confirming conjectures in the lit-

erature. In so doing, we have found that the energy computed from a non-self-consistent Kohn-Sham calculation using the TFW ground state density as input is in very good agreement with the energy obtained from the fully self-consistent Kohn-Sham solution.

The development of more general and accurate electronic kinetic energy functionals for use in orbital-free DFT, possibly aided by state-of-the-art machine learning techniques, is therefore a worthy subject of pursuit.

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