

Comment on “Accurate Correlation Potentials from the Self-Consistent Random Phase Approximation”

Trushin *et al.* [1] developed a self-consistent random phase approximation, which could yield highly accurate exchange-correlation potentials and electron densities. However, we argue that the metric they used to assess the error of the density is insufficient, because it leads to the conclusion that the Hartree-Fock (HF) density is superior to PBE and r^2 SCAN densities—a claim that, while aligned with density-corrected density functional theory [2], is inconsistent with evidence presented here and in recent studies [3]. Those studies show that the higher energy accuracy of density functional approximations (DFAs) when evaluated using the Hartree-Fock density is not necessarily because the HF density is more accurate but often due to a cancellation of functional-driven error from DFA by density-driven error from the HF density.

In Fig. 3 in the main text in Ref. [1], as in our Fig. 1(a), the absolute density difference relative to the CCSD(T) density of the CO molecule is plotted along the bond axis. We used the PySCF code [4] with the aug-cc-pwcvqz basis set (except where noted otherwise). The results show that PBE and r^2 SCAN exhibit significant error near the nuclei, while the HF density error is larger in regions farther from the nuclei. This outcome is expected, as PBE and r^2 SCAN suffer from substantial self-interaction errors in the core region, whereas HF is free of self-interaction. The full Perdew-Zunger self-interaction correction mitigates this error, as shown in Fig. 1(b), where the self-interaction corrected r^2 SCAN and HF exhibit similar density errors at the nuclei. We argue that the density along the bond axis is not representative of the overall electron density. A more appropriate metric is the projected density difference, defined as

$$\Delta\rho(z) = \int \int dx dy |\rho - \rho^{\text{CCSD(T)}}|. \quad (1)$$

In Fig. 1(c), the projected density difference is plotted along the bond axis, demonstrating systematic improvement from HF to PBE to r^2 SCAN. This trend is consistent with the computed dipole moments of the CO molecule: The Hartree-Fock density yields a dipole moment of -0.2645 D, which has an incorrect sign. In contrast, PBE and r^2 SCAN produce dipole moments of 0.2245 and 0.1681 D, respectively, in improving agreement with the CCSD(T) value of 0.1172 D [6]. Our conclusions about

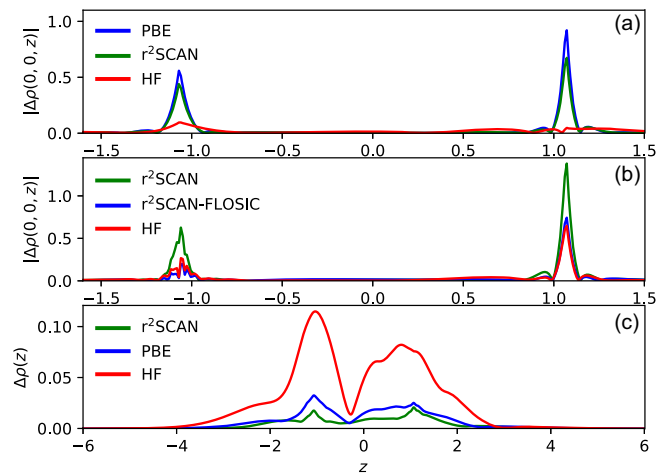


FIG. 1. (a),(b) Absolute density difference along the CO bond axis (C on the left, O on the right). In (b), the NRLMOL basis set was used for r^2 SCAN, r^2 SCAN-FLOSIC, and HF. r^2 SCAN-FLOSIC was computed with the FLOSIC code [5]. (c) Projected density along the CO bond axis. The unrelaxed CCSD(T) density serves as reference in all panels.

the HF and density-functional electron density of *sp* molecules also agree with those of Refs. [7,8].

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Data availability—The data that support the findings of this article are openly available [9].

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