CAS-DFT based on Coulomb-division scheme

S. Yamanaka[a], T. Ukai[b], K. Nakata[b], K. Kusakabe[c],
Y. Yonezawa[d], H. Nakamura[d], T. Takada[e], and K. Yamaguchi[a]

[a] Center for Quantum Science and Technology under Extreme Conditions, Osaka University, Toyonaka, 560-8531, Japan.

[b] Department of Chemistry, Graduate School of Science, Osaka University, Toyonaka, 560-0043, Japan.

[c] Department of Physical Science, Graduate School of Engineering Science, Osaka University, Toyonaka, 560-8531, Japan

[d] Institute for Protein Research, Osaka University, Toyonaka, 560-8531, Japan

[e] RIKEN, Next-Generation Supercomputer R&D center, Chiyodaku, Tokyo, 100-0005, Japan

During the past decade, a new class of electronic structure theory combining multireference wavefunction and density functional (MR-DFT) has been developed to cover the systems that exhibit strong multireference character [1-5]. There are various types of MR-DFT, which are classified into Coulomb-driven [1-3] and wavefunction-driven ones [3-5]. The former is based on the division of electron repulsion Coulomb operator into short-and-long range parts. On the other hand, wavefunction-driven MR-DFT is based on truncation of wavefunction expansions, and remaining correlations are covered by DFT. From the computational viewpoint, a modest wavefunction (WF), such as complete-active-space (CAS) WF, is desirable. Indeed, we present CASCI/CASSCF-DFT methods that satisfy the variational principle for both ground and excited states [3,5]. In these previous studies, we employ MSS-GC scheme [4] for CAS-DFT. In this study, we propose a new type of MR-DFT, in which a long or short-range part of the Coulomb repulsions for nondynamical correlations is covered by CAS wavefunction and other all parts are covered by DFT. Numerical examples are presented for both ground-and-excited states of simple systems.

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