An effective grid-free approach based on the third-generation density-functional-theory calculation method

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We have developed the third-generation (3G) density-functional-theory (DFT) based calculation method [1] to calculate the standard canonical molecular orbitals (CMOs) of giant molecular system such as proteins. The 3G DFT method is based on modified Cholesky decomposition (CD) to evaluate the Coulomb and exact exchange terms, and the grid-free method to do the exchange-correlation (XC) term. The Fock (Kohn-Sham) matrix is gained by only simple matrix-matrix operations in the SCF loop, so the method is a technology designed to permit massively distributed-memory parallel computers to be efficiently used for the CMOs calculation of giant system.

In this report, we demonstrate a modified grid-free approach employing the XC term in the 3G DFT framework. The original grid-free approach analytically provides the XC term by a matrix representation of density (MRD) [2]. The previously reported approach had used a density-fitting method for minimizing computational costs, requiring auxiliary basis in advance. We indicated that the basis set of the MRD acts a drawback, and demonstrate a practical improvement. Additionally, we propose an effective computation based on the 3G DFT method under the effective parallel computation and error control.

This method is implemented to our DFT program, ProteinDF, which is distributed under the GPL v3 via GitHub (https://proteindf.github.io/).

[1] TH and FS, *Phys. Chem. Chem. Phys.*, **16**, 14496 (2014).
[2] Y. C. Zheng and J. Almlöf, *J. Chem. Phys. Lett.*, **214**, 397 (1993).