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Comment

Comment on "A generalized fast multipole approach for Hartree–Fock and density functional computations"

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Abstract

Two approaches for forming the J-matrix of ab initio density functional theory calculations in time scaling linearly with molecular size are compared in terms of accuracy and efficiency.

A major step in large scale ab initio density functional theory (DFT) calculations using atom-centered Gaussian basis functions is formation of the J-matrix, corresponding to averaged Coulomb interactions between electrons. We recently presented the first method for constructing the J-matrix in time which can scale linearly with the size of the molecule [1]. Our method, termed the continuous fast multipole method (CFMM) [1], is a generalization of the fast multipole method (FMM) [1]. The CFMM generalizes the FMM by including the intrinsically nonzero extent of the distributions associated with products of basis functions in a manner that retains the desirable error bound of the FMM for point charges. For systems where the extent of charge distributions is small relative to the size of the molecule, linear scaling is obtained. The FMM is exactly recovered in the limit where all distributions have negligible extent, while for systems where all distributions overlap, all interactions must be calculated explicitly, and no improvement in scaling can be attained.

Kutteh, Aprà and Nichols (KAN) [3] have subsequently suggested an alternative approach to the J-matrix problem, which they conclude is 'more efficient', 'more straightforward', and 'more general', than the CFMM. They further suggest the CFMM is not only 'more obvious', but also has a 'restricted range of applicability'. The KAN approach involves direct application of the point charge FMM to the J-matrix problem. Overlapping charge distributions are circumvented by decomposing the charge density into a set of atom-centered multipoles, as the input to a conventional FMM.

We believe that KAN have probably achieved a more efficient method than the CFMM, but one which is also *drastically less accurate*, an issue they

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[1] For an overview of the FMM and related methods, see Greengard [2].

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have failed to consider in the comparison of the two methods [3]. To address this key consideration in more detail, we note the following points:

(1) A central feature of the CFMM, which is retention of a well-defined error bound by properly treating the extent of charge distributions, is sacrificed in the KAN alternative due to the initial decomposition of the density into local multipoles. This may well make the KAN method more efficient, but the efficiency comes at the price of *unpredictable* errors whose magnitude will have to be established empirically. As an extreme example, the KAN method achieves linear scaling in the limiting case where all distributions overlap, and multipole representations are not convergent!

(2) Subsequent steps in the KAN method involve bounded errors. However, KAN advocated a low-order implementation of their method using the Cartesian FMM, by contrast with the CFMM, which employs spherical harmonics of any specified order. This gives the KAN method greater efficiency when low-order Cartesian multipoles are compared against high-order spherical multipoles in the CFMM, but again at the price of increased levels of error. For a *given* multipole truncation, the spherical harmonic method requires fewer floating point operations.

(3) Based on the stated parameters of KAN [3], we can estimate the error from point (2), using CFMM calculations [1]. Errors on the order of millihartrees are obtained by truncating multipole expansions at the hexadecapole level for beyond next-nearest neighbor ($ws = 2$) interactions as advocated by KAN. By contrast, the CFMM with $ws = 2$, truncating at 10-poles, yields sub-microhartree errors, as necessary to permit accurate reproduction of 'exact' calculations of the J-matrix. Thus, separate from the unbounded errors arising from point (1), chemically significant errors arise from the use of low-order multipole expansions.

(4) Unlike the CFMM, and despite its touted simplicity, the KAN algorithm was reported without implementation [3]. This is a regrettable practice, particularly for a numerical algorithm without bounded errors. We await full comparison with our latest results [1,4] with interest.

References

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