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The continuous fast multipole method

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Abstract

We introduce the continuous fast multipole method (CFMM), a generalization of the fast multipole method for calculating Coulomb interaction of point charges. The CFMM calculates Coulomb interactions between charge distributions, represented by continuous functions, in work scaling linearly with their number for constant density systems. Model calculations suggest that for errors in the potential of 10^{-10} , the CFMM becomes faster than direct evaluation for less than 10000 Gaussian charge distributions. Using the CFMM to form the J matrix in ab initio density functional and Hartree–Fock calculations shows that a two–three times speedup is attainable for the linear alkanes $C_{10}H_{22}$ – $C_{20}H_{42}$.

1. Introduction

The fast multipole method [1] (FMM) calculates the Coulomb interaction between classical point charges in work which scales linearly with the number of particles in the system ¹. It has been shown to reduce the time involved in calculating the potential for a collection of discrete point charges relative to direct pairwise evaluation, with crossovers on the order of thousands of particles [3–9]. However, the restriction to point charges means the FMM is not immediately applicable to problems in which the charge distributions have significant extent.

Ab initio electronic structure calculations constitute such a class of problems. For the largest molecular systems whose study is currently feasible by Hartree–Fock [10] and density functional theory [11], the rate-limiting step is computing the electro-

static interactions between all pairs of electrons in the system. These electrons are often represented by local basis functions such as Gaussians [12]. Over the last two decades, a variety of computational strategies have been suggested and implemented for treating this problem [13–21]. While these methods have resulted in tremendously improved efficiency, they all scale quadratically with the number of basis functions.

To realistically treat large molecules, the quadratic scaling must be reduced. For this reason, we have generalized Greengard and Rokhlin's FMM to treat these continuous distributions. The classical point charge FMM [1–9] systematically organizes multipole representations of local charge distributions, so that each particle interacts with local expansions of the potential due to all distant particles. To group the particles, they are placed in a box which is repeatedly subdivided to create local collections of point charges. Local and distant distributions are distinguished by the global well-separated (WS) index [3]. This is de-

¹ For a recent general audience review of fast-summation algorithms see Ref. [2].

defined as the number of boxes which must separate two collections of charges before they may be considered distant, and can interact through multipole expansions.

The above prescription fails for continuous distributions because in this case, a single distribution can cover the entire original box. This makes the straightforward division of this box to create individual local collections impossible. To alleviate this problem, we treat distributions having various extents differently in the method presented here, which we term the continuous fast multipole method (CFMM). A distribution having a small extent relative to the total extent of the system must have a different definition of local and distant (WS index) than a distribution of large extent.

In the CFMM we generalize the WS index and reorganize the sorting of the distributions. We can define a unique center for each distribution, place these centers in a box, and subdivide this box to create local collections of centers of distributions. To define distant and local, we give *each* distribution its own WS index. The distributions are sorted by their WS indices as well as the positions of their centers in space, to properly define local versus distant interactions.

This generalization will give linear scaling only in the case where the density of distributions in the system grows in a sub-linear manner with increased numbers of distributions. More simply, as one adds distributions to the system, one must increase the total volume of the system (as measured by a box enclosing the centers of the distributions). In a case of greater than linear behavior asymptotically all of the distributions must overlap. In this case, there is no classical behavior and the CFMM becomes a fully quadratic method.

This paper gives a concise description of the generalization of FMM to create the CFMM (Sections 2 and 3) and a numerical exploration of the error and timing issues involved in its use (Sections 4 and 5). Section 2 describes the specific modifications needed, while Section 3 summarizes the complete CFMM algorithm. In Section 4 we explore the errors and timing issues for the use of the CFMM for a simple model system. Section 5 examines the application of the CFMM to the calculation of the J matrix typically used in Hartree–Fock, and density functional calcu-

lations for a series of linear alkanes. Finally the Appendix gives a simple method for the formation of the multipole integrals of continuous functions.

2. Well separatedness of local distributions

We define the extent of a distribution such that two distributions separated by the sum of their extents will interact as classical point charges to a desired precision [21]. The mathematical expression for the extent of a distribution will depend upon the functional form of the distribution. Most ab initio electronic structure calculations use Gaussians as the basis for representing quantum mechanical charge distributions [12]. For this reason, we will concentrate mainly on the application of the CFMM to Gaussian distributions, although the formalism presented is valid for any localized continuous distribution where a definition of an extent is possible.

The mathematical definition for the extent of a Gaussian distribution is exceptionally simple. The Coulombic interaction of two spherical Gaussian charge distributions can be represented in closed form as

$$V = \frac{1}{|\mathbf{R}|} \operatorname{erf} \left(\sqrt{\frac{pq}{p+q}} |\mathbf{R}| \right), \quad (1)$$

where \mathbf{R} is a vector connecting the centers of the distributions and p and q are the exponents of the Gaussians. The factor $\operatorname{erf}(\sqrt{pq/(p+q)} |\mathbf{R}|)$ rapidly approaches 1 with increasing separation, and the two distributions then interact as classical point charges. Consequently, we define the extent of a distribution as

$$r_{\text{ext}} = \sqrt{\frac{2}{p}} \operatorname{erf}^{-1}(1 - \epsilon), \quad (2)$$

where p is the exponent of the Gaussian, and ϵ is the desired precision. For higher angular momentum functions, the expression is similar with the classical limit being interactions of higher-order multipole moments. The purpose of the CFMM is to group distributions in such a manner so as to know which distributions can be treated classically through multipole expansions.

The base well separatedness (WS_{ref}) index governs

the error obtained in a CFMM calculation. Distributions will not be treated through the multipole expansion if their centers are closer than the base WS index. For example $WS=2$ means at least two boxes must separate the boxes enclosing the interacting distributions for multipoles to be used. Since the distributions have non-zero extent, it is possible for the centers of two distributions to be well separated as defined by WS_{ref} , and yet still be unable to interact classically. The actual WS index for each distribution used by the CFMM must reflect the true extent (as defined in Eq. (2) for Gaussians) of the distribution. We define the WS index for each distribution as

$$WS = \max \left(2 \left\lceil \frac{r_{ext}}{l} \right\rceil, WS_{ref} \right), \quad (3)$$

where l is the box size (which depends on the level of the tree). Distributions having the same WS index will be grouped together forming a branch of the CFMM tree structure (Fig. 1). In parts of the CFMM algorithm where information is passed up the tree (the first pass of Section 3), the fact that the box size doubles means that from Eq. (3), the WS index for each distribution will be halved until WS_{ref} is reached.

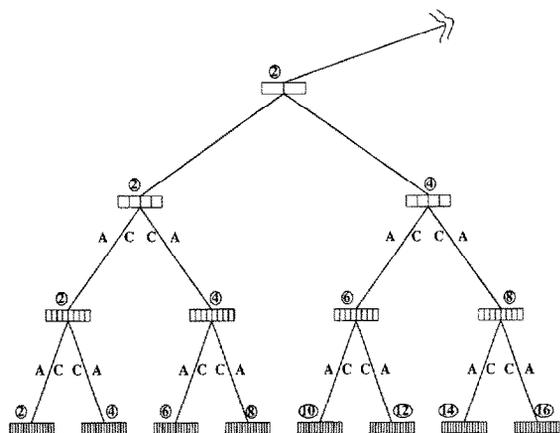


Fig. 1. A 1D representation of the CFMM tree structure with a base WS index of 2. Each level of the tree divides space into 2^{level} boxes (8^{level} in 3D). These boxes are used to sort the distributions by the locations of their centers. Each level contains several branches, each branch containing all distributions having the same WS index (indicated by the encircled number). As multipole/Taylor information is shifted up (A) and down (C) the tree [1–9], the WS index for a given branch may be reduced/increased by a factor of 2. This allows the sharing of multipole/Taylor information, and gives rise to the binary-tree structure.

3. The continuous fast multipole method

3.1. Pass 1: form and translate multipoles

The continuous fast multipole method closely resembles the point charge version we have described previously [3]. In the CFMM we have multiple branches of boxes on the lowest level of the tree (Fig. 1) each corresponding to a different WS index. We radix sort the distributions into the lowest level boxes by the spatial location of their centers, and by their extents. The distributions which cannot interact with any other distributions via multipoles are placed within a special level which only interacts directly. The multipole moments of all distributions centered within a lowest level box are formed about the center of that box. These moments are formed analytically by the method described in the Appendix. As with the FMM these moments are translated up the tree. The translations follow the pathways marked A in Fig. 1. As discussed above, each translation up reduces WS by a factor of 2, which as shown in Fig. 1 allows the information contained in branches with different WS indices to be combined on higher levels.

3.2. Pass 2: convert multipoles to Taylor expansions

Pass 2 of the FMM algorithm converts multipole expansions centered in one box into local Taylor expansions about the center of another box. This conversion happens only for well-separated boxes. In the CFMM this portion of the algorithm is most altered from the classical point charge FMM. Pass 2 works independently upon each level of the tree. Each level is composed of several branches each containing distributions having different WS indices. Within a given branch, pass 2 behaves exactly like the FMM pass 2 for a single WS value. *Between* branches pass 2 uses the average of the WS values for each branch to determine whether particular boxes are well separated. For example, translation between boxes from $WS=2$ and $WS=4$ branches of a given level will only occur if there are more than three intervening boxes (unless the parent boxes are well separated in which case the translation occurs at the parent level). This procedure ensures the maximal number of distributions contained in the two branches can interact classically via multipoles.

3.3. Pass 3: translate Taylor information to lowest level

The local Taylor expansions created on higher levels of the tree in pass 2 must now be translated to the lowest level boxes. We now translate from each parent to all children of that parent (paths labeled with C in Fig. 1). At the end of this pass, we have within each of the lowest level boxes, a Taylor expansion representing the potential from all well-separated distributions.

3.4. Pass 4: evaluate Taylor expansions to obtain far-field potential

Pass 4 is identical to the point charge version. The Taylor expansion in each lowest-level box represents the potential due to all well-separated distributions. We can evaluate this expansion at points of interest (i.e. integrating over each distribution) in each lowest level box to produce a far field potential for each distribution.

3.5. Pass 5: perform direct interactions between overlapping distributions

The direct interactions of charge distributions require the evaluation of the standard Coulomb integrals. For each distribution, we perform direct interactions with all particles located in boxes which are not well-separated producing a near-field potential. This determination is straightforward given the initial assignment of distributions to the lowest level branches of the tree in pass 1, and the averaging procedure discussed in pass 2 for distributions on different branches. This potential is added to the far field potential obtained in pass 4 to produce the total potential for each distribution.

4. Random model system

In the limiting case where all of the distributions in the system have negligible extent in comparison with the total size of the system, the CFMM reduces to the FMM with a WS value given by WS_{ref} as will the errors and timings. Likewise, when all of the distributions have a common WS value greater than WS_{ref}

the errors and timings should be governed by the FMM estimates for that WS value. Only in the case where distributions have differing extents, and thus different WS values, will one see behavior unique to the CFMM.

We have chosen a model system of the latter type, which is loosely related to a random distribution of Coulombically interacting hydrogen atoms. As we add hydrogens to the system, the volume of the enclosing box is increased to keep the density at 1 hydrogen/ \AA^3 ². Each hydrogen atom has a positive unit charge nucleus represented by a Gaussian with a large exponent and a collection of ten Gaussians with various exponents representing the negative electron. The Gaussians representing the electron are randomly centered throughout the system³ with coefficients selected to sum to a negative unit charge thus making the system globally neutral.

The exponents of the Gaussians have been randomly chosen to range between 0.1 and 20 to model a regime appropriate for electronic structure calculations. Small exponents less than 3 will show poor CFMM performance for small systems while large exponents greater than 20 will show results similar to the point charge FMM. As the systems become larger these differences become less pronounced.

The base WS index governs the upper bound on the error in the CFMM. However in the case where several WS values are present, this estimate is very conservative. If a large number of the distributions are located on high WS index branches, we should see an error much less than the FMM error estimate. In this section we present numerical results for our model system showing the growth of the error caused by truncation of the multipole expansion and increasing the number of particles in the system.

Fig. 2 shows the error as a function of the level of multipoles used in the calculation. The plot shows calculations on the model system with 10000 distri-

² It should be noted that in the case where the density of the system grows at a rate greater than linear with the number of distributions, we are effectively placing the distributions directly on top of each other. In this case, CFMM *must* asymptotically scale *quadratically*. The worst case is all of the distributions centered about the same point.

³ This is to ensure that each hydrogen of the system is not individually neutral which would make all long range interactions zero.

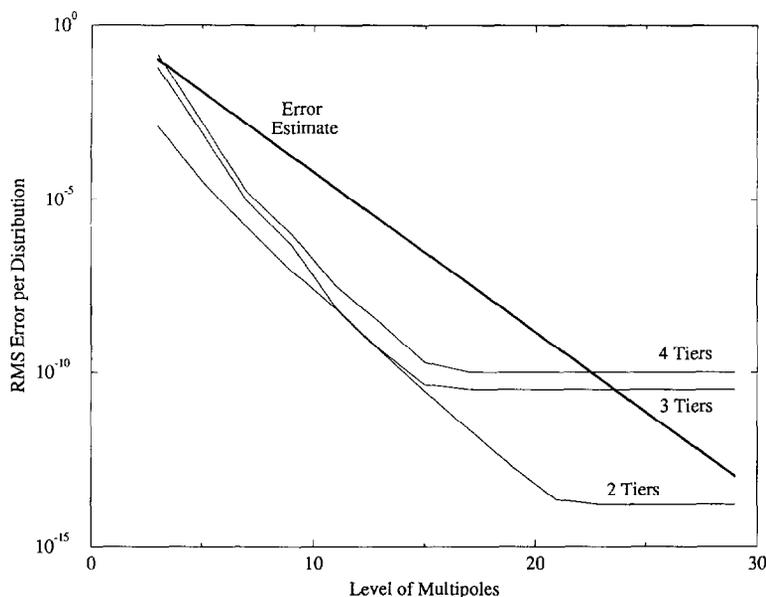


Fig. 2. The RMS error per distribution for a 10000 distribution system as a function of the number of multipoles used in the calculation. The errors indicate that to obtain the potential to 10^{-10} , one need only use 15 multipoles. Several tree depths are displayed indicating that the size of the boxes on the lowest level of the tree also enters the CFMM error expression. The error estimate for a standard FMM calculation (2 tiers; $WS=2$) is also shown on the graph indicating that using the CFMM base WS index gives very conservative estimates of the CFMM error.

butions using various depths for the CFMM tree. At approximately 15 multipoles for all depths, we reach a point where error becomes less than 10^{-10} . The FMM error estimate for the base WS index and a 2 tiered tree is also shown on the figure indicating the conservative nature of this estimate for the CFMM calculation.

The error estimates for the point charge FMM indicate a growth in the absolute error with the number of particles treated. Fig. 3 numerically explores the growth in the CFMM error with the number of distributions. The figure shows the sub-linear growth in CFMM errors with the number of particles for a fixed level of multipoles. This coupled with the information contained in Fig. 2 indicates that to achieve machine precision, one need only use between 20 and 30 multipoles.

All of the timings presented have been obtained on a DEC ALPHA/AXP 3000/600 workstation running OSF/1. Fig. 4 shows the time for a CFMM calculation as a function of the number of distributions. In this graph we show the minimum time for treating systems with 10 and 20 multipoles (the errors ob-

tained for most of these calculations are shown in Fig. 3). It is always possible to determine whether interacting two boxes directly will be faster than interacting the boxes via multipoles: our implementation of the CFMM defaults to direct interactions in these cases. This causes the crossovers to be well under 10000 distributions.

5. Density functional theory: J matrix formation

Electronic structure calculations directly benefit from the favorable scaling properties of the CFMM. In both density functional theory (DFT) and conventional Hartree-Fock (HF) theory, one requires the evaluation of the so-called J matrix. $J_{\mu\nu}$ represents the Coulomb interaction of a given distribution (formed by the overlap of two basis functions, $\varphi_{\mu}(\mathbf{r})$ and $\varphi_{\nu}(\mathbf{r})$) with the molecular electron density, which is expanded in terms of all other distributions in the molecule:

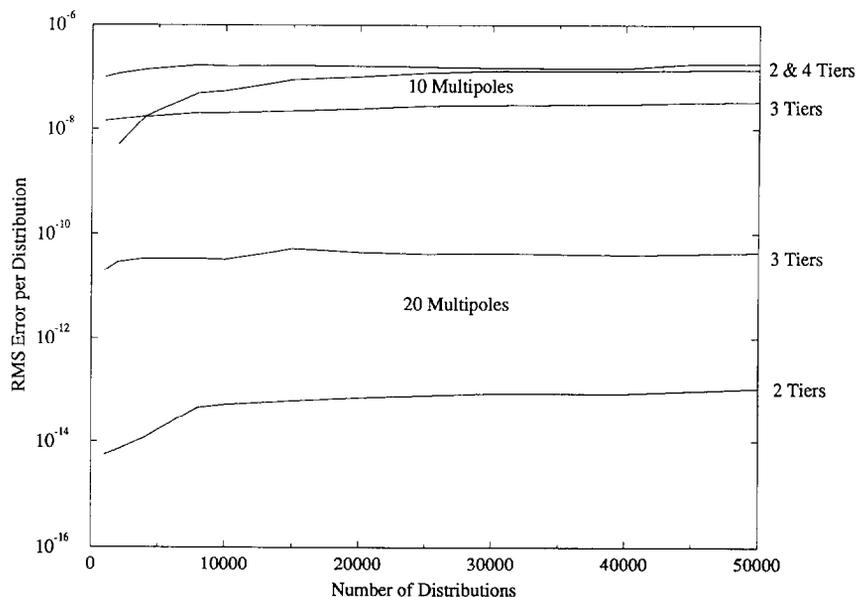


Fig. 3. This figure numerically explores the growth of the CFMM error with the number of distributions treated. Calculations using 10 and 20 multipoles are displayed showing similar sub-linear behavior in the growth of the errors with the number of distributions. The timings for the calculations portrayed in this figure are given in Fig. 4.

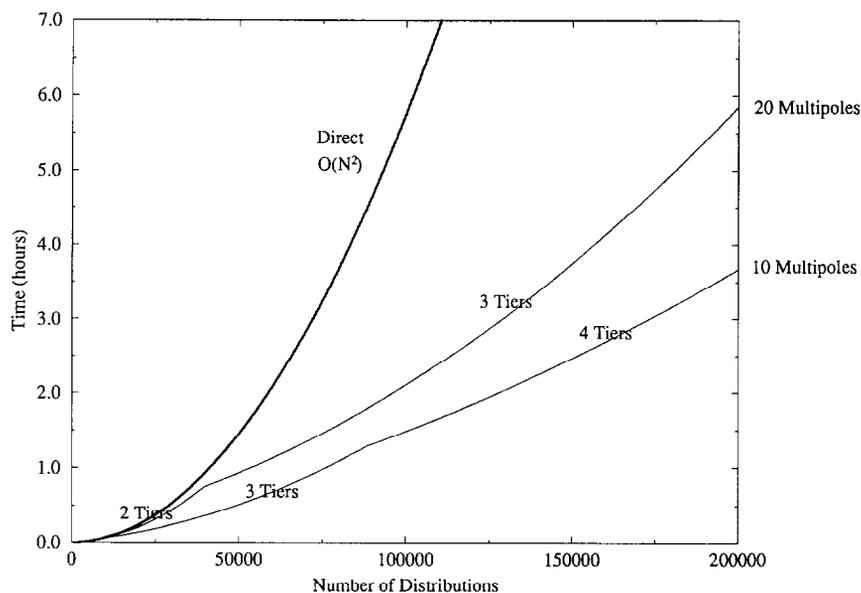


Fig. 4. The time to perform a CFMM calculation as a function of the number of distributions contained in our simple model system. The shallow cusp-like behavior characteristic of fast multipole methods [3] can be seen as one increases the depth of the CFMM tree. Fig. 3 can be used to assess the accuracy for a given number of multipoles and tier depth for the timings shown in this figure. The cross-overs for both levels of multipoles are obscured due to the scale of the graph; they occur at less than 10000 distributions with the CFMM calculation never more than 5 min longer than the direct calculation.

$$J_{\mu\nu} = \iint d\mathbf{r}_1 d\mathbf{r}_2 \varphi_\mu(\mathbf{r}_1) \varphi_\nu(\mathbf{r}_1) \frac{1}{r_{12}} \times \left(\sum_{\lambda\sigma} P_{\lambda\sigma} \varphi_\lambda(\mathbf{r}_2) \varphi_\sigma(\mathbf{r}_2) \right). \quad (4)$$

The CFMM code is currently being interfaced with the Q-Chem [22] computational chemistry package. While the preliminary interface is not optimized, it does allow the CFMM to calculate the J matrix in DFT [23]. This permits an initial assessment of the usefulness of the CFMM for treating real molecules with standard basis sets including non-spherically symmetric functions. Due to the preliminary nature of the interface, we will not present timings within this communication. However, we can report the number of interactions saved since this value will not change with changes in the interface.

Our DFT calculations are on a series of linear alkanes using the standard STO-3G basis [24]. For all of the calculations, we have used 25 multipoles and have obtained energies correct to at least ten significant figures. The distributions are products of pairs of basis functions. A pre-screening process with a threshold of 10^{-10} was used to remove the negligible distributions. We find the number of distributions asymptotically grows linearly with the size of the molecule.

The number of interactions saved for several molecules is reported in Table 1. In practice for trees less than 5 tiers deep, the amount of time spent performing passes 1–4 is vastly overshadowed by the time spent performing the direct interactions in pass 5. Thus we can roughly associate the % of interactions saved with the amount of time saved in the calculation. Using this measure, we find that for this series of molecules we obtain a two times speedup over conventional methods by $C_{10}H_{22}$ and a three times speedup by $C_{25}H_{52}$.

As a final example, to gain insight into the potential of the CFMM for very large molecules, we have examined a small fragment of DNA, the six base-pair oligonucleotide CTCGAG, which has 378 atoms. This molecule is beyond the memory limitations of our current interface, however, we have performed several hypothetical calculations (Table 2) which count the number of interactions which must be done directly and via multipoles. The results indicate for

Table 1
 J matrix formation (10^{-10} threshold). CFMM/DFT calculations on the homologous series of linear alkanes using the STO-3G basis [24]

System	Basis functions	Distributions	% long-range interactions
three-level tree			
C_3H_{12}	37	2675	19
$C_{10}H_{22}$	72	6767	32
$C_{15}H_{32}$	107	10862	41
$C_{20}H_{42}$	142	14957	43
$C_{25}H_{52}$	177	19052	44
four-level tree			
C_3H_{12}	37	2675	29
$C_{10}H_{22}$	72	6767	49
$C_{15}H_{32}$	107	10862	59
$C_{20}H_{42}$	142	14957	63
$C_{25}H_{52}$	177	19052	67

All calculations are performed using 25 multipoles to obtain energies correct to at least 10 significant figures. The distributions reported arise as overlapping basis functions, a pre-screening process enabled linear growth of the number of distributions with increasing molecular size. These results indicate a two–three times increase in speed of a CFMM calculation over a conventional direct implementation.

Table 2
Characterization of interactions and WS definitions in CTCGAG STO-3G at various tree depths

Depth	% long-range interactions	Highest WS value	% distributions at WS 2
3	66.4	2	100.0
4	87.0	4	79.5
5	93.4	8	52.0
6	95.3	16	21.0

The percentage of long-range interactions in the CTCGAG J matrix calculation using the STO-3G [24] basis for various depths of the CFMM tree. The system has 1370 basis functions, and approximately 128000 distributions at the single-point accuracy used. These results indicate almost an order of magnitude increase in the speed of calculation as compared to direct methods.

molecules of this size, we can expect a speedup of almost an order of magnitude.

6. Conclusions

The main results and conclusions of this paper are

as follows:

(1) We have introduced the continuous fast multipole method (CFMM) as a generalization of Greengard's point charge FMM to treat Coulomb interactions of local charge distributions represented by continuous functions. In the FMM, long range Coulomb interactions are efficiently handled by multipole and local expansions. The CFMM organizes distributions having different extents into a computational hierarchy which permits potential evaluation to scale only linearly with system size.

(2) Numerical tests of the CFMM suggest that a manageable number of multipoles (≈ 25) will provide results accurate to better than 10^{-10} . The errors involved have also been shown to grow only weakly with the number of distributions.

(3) At 10^{-10} accuracy, timings show crossovers in the tens of thousands of distributions for systems having exponents ranging from 0.1 to 20. This is a fairly small degradation relative to the point charge limit, and indicates that the CFMM may improve calculational speed for a wide variety of problems.

(4) One such problem is the interelectronic Coulomb interaction in density functional theory (DFT) calculations of molecular electronic structure. Preliminary DFT studies show the CFMM has great potential for the calculation of the J matrix in large molecules. The number of interactions saved show a possible two–three times increase in speed for calculations for the series of linear alkanes. This factor will increase as we begin to treat larger and larger molecules. The CFMM may complement 'divide and conquer' methods [25–27] which propose the diagonalization of an effective Hamiltonian in linear time.

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Appendix: Efficient formation of multipole moments

The multipole moments of charge q_a located at \mathbf{a} (α, β) with respect to a local origin \mathbf{o} are defined by

$$\omega_{lm}(\mathbf{a}) = q_a \cdot O_{lm}(\mathbf{a}) = q_a a^l \tilde{P}_{lm}(\cos \alpha) e^{-im\beta}. \quad (\text{A.1})$$

The multipole moments of a charge *distribution* about a given origin are the summation of the individual multipole moments for each charge in the distribution about the same origin. The evaluation of the multipole moments of a continuous distribution requires an integration of the above expression over the entire distribution $\chi(\mathbf{r})$ centered at \mathbf{r}' ,

$$\omega_{lm}(\mathbf{r}') = \int d\mathbf{r} \chi(\mathbf{r}' - \mathbf{r}) \cdot O_{lm}(\mathbf{o} - \mathbf{r}). \quad (\text{A.2})$$

Notice this expression simplifies considerably if the origin \mathbf{o} of the multipole expansion coincides with \mathbf{r}' the origin of the charge distribution. In this case

$$\begin{aligned} \omega_{lm}(\mathbf{r}') &= \int d\mathbf{r} \chi(\mathbf{r}' - \mathbf{r}) \cdot O_{lm}(\mathbf{r}' - \mathbf{r}) \\ &= \int d\mathbf{r} \chi(\mathbf{r}) \cdot O_{lm}(\mathbf{r}). \end{aligned} \quad (\text{A.3})$$

This leaves a simple integral over $\chi(\mathbf{r})$. We may now use the symmetry properties of $\chi(\mathbf{r})$ to reduce the number of moments we need to calculate. For the cases we will examine, this expression can be determined analytically. We can now translate these moments in space to produce moments about our original origin \mathbf{o} ,

$$\omega_{lm}(\mathbf{o}) = \sum_{j=0}^l \sum_{k=-j}^j A_{jk}^{lm}(\mathbf{o} - \mathbf{r}') \cdot \omega_{jk}(\mathbf{r}'). \quad (\text{A.4})$$

The translation is performed using the operator $A_{jk}^{lm}(\mathbf{a}) = O_{l-j, m-k}(\mathbf{a})$ developed for use in the fast multipole algorithm [3].

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