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Technical Report

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A Particle-Particle Particle-Multigrid Method for Long-Range Interactions in Molecular Simulations

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Abstract: A new fast method of order $\mathcal{O}(N)$ is proposed to calculate interaction energies and forces in molecular systems with open boundaries, exerted by long range Coulomb interactions. The method consists of a fast multigrid Poisson solver for the far field smooth part of the potential and a particle-particle based method for the near field contribution. Boundary conditions are calculated with a multipole expansion method. Test cases are conducted for performance measurements of the method.

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1 Introduction

Coulomb interactions often play a crucial role for static and dynamical properties in a variety of complex systems, characterized by polar or charged system componends, e.g. polar liquids, proteins, DNA, membranes, polyelectrolytes or plasmas. Due to the long range nature of these interactions their determination is computationally very demanding. Since pair interactions of all particles in the system have to be taken into account, the problem has intrinsic complexity $\mathcal{O}(N^2)$. Therefore the size of the systems or length of the system trajectory is mainly limited by the computational overhead induced by electrostatic interactions. Due to the great interest in systems, dominated by Coulomb interactions, there was great effort spent in developing faster and more efficient methods with a lower complexity. For systems with periodic boundary conditions the most widely used method is the Ewald summation technique which is formally an exact analogue of an infinite lattice sum. Practically a small, controlable truncation error is accepted and it could be shown that for a given error, the method scales like $\mathcal{O}(N^{3/2})$ [1]. A faster variant of this method was later on developed, using fast Fourier transform techniques to reduce the complexity to $\mathcal{O}(N \log(N))$ [2]. A modification of the Ewald summation is the so called particle-particle particle-mesh method ($\mathrm{P^3M}$) which solves the field equation with a fast Fourier method, using a modified Green's function, which adjusts the solution closely to the continuum solution. The method splits the field into

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near and far field contributions, where the short range part is calculated by an explicit particle-particle summation. Due to the fast Fourier transform the complexity is again reduced to $\mathcal{O}(N\log(N))$ [3].

For open systems, mainly two fast methods are in use, which both profit from a multipole expansion of far field contributions of charges to the local field. The Barnes-Hut tree algorithm splits the contributions hierarchically to end up in an $\mathcal{O}(N\log(N))$ complexity [4], while the Fast Multipole Method (FMM) reduces this complexity to $\mathcal{O}(N)$ by taking into account interactions of multipoles [5].

In the present article a new method for open systems is proposed which goes in line with the idea of the P^3M method, i.e. the near- and far-field contributions are treated in seperate ways. The idea here is to apply the multigrid (MG) method to the solution of Poisson's equation to calculate a global potential energy surface. Normally, this problem would consist in an $\mathcal{O}(N^2)$ complexity. However, multigrid methods reduce this complexity to $\mathcal{O}(N)$, making them very attractive to many body problems. Having the global solution it is required to correct for the self energy of the particles and the contributions from the near field part. This is performed via subtracting the grid based Green's function. In a last step the near field part is calculated as a pair-sum over all neigbored particles. In the following the mathematical basis is shown and performance measurements are compared with explicit pair-wise calculations.

2 Theory

In this section a description of the different steps, involved into the calculation of the interaction energies and forces is given.

Multigrid Solution of the Poisson Equation

Consider the Poisson equation for the potential energy Φ, subject to Dirichlet boundary conditions,

$$\Delta\Phi(\mathbf{r}) = -\rho(\mathbf{r}) \quad , \quad \mathbf{r}\in\Omega \tag{1}$$

$$\Phi(\mathbf{r}) = \Phi^s(\mathbf{r}) \quad , \quad \mathbf{r}\in\partial\Omega \tag{2}$$

where ρ is the charge density and $\Phi^s(\mathbf{r})$ the *known* field, created by the N charges on the surface of the system. To apply multigrid techniques the equation is discretized, i.e. the Laplace operator translates into a finite difference approximation, $\Delta \to D_h$, where h indicates the mesh spacing. The charges in the system are smeared onto the grid points via Cloud In Cell (CIC) weighting functions w [3], resulting in the grid based charge density

$$\hat{\rho}_h = \frac{1}{h_x h_y h_z}\sum_{i\in C(h)} q_i \prod_{\alpha=x,y,z}\left(1-\frac{|\delta r_{i,\alpha}|}{h_\alpha}\right) \tag{3}$$

where $C(h)$ are those cells which share grid point h and $\delta r_{i,\alpha}$ are relative particle coordinates with respect to grid point h (in the following grid based functions

are indicated with "$\hat{}$"). The idea of multigrid [6] is now to solve the Poisson equation on a hierarchy of fine and coarse grids with finite difference schemes. In the present work a 4th-order compact solver is used [7] in combination with a Gauss-Seidel relaxation scheme. On the finest grid the operator D_h works on the field $\hat{\Phi}_h$, giving an approximation $\hat{\Phi}'_h$. It can be shown [6] that the resulting error $\hat{\epsilon}_h$ with respect to the exact solution itself obeys a Poisson equation, $D_h\hat{\epsilon}_h = -\hat{r}_h$, where the source term consists of the residuum left in the relaxation step. Solving for the error gives therefore a correction to the first approximation. The error correction is done on the next coarser grid with mesh size $H = 2h$, where the residuum is restricted from $h \to H$ via a restriction operator, $\hat{r}_H = I_h^H \hat{r}_h$. It is found that the high frequency components of the error are rapidly removed on a given grid. Therefore coarsening the grid results in the fact that low fequency components from the fine grid are transformed into high frequency components on the coarse grid, which can be efficiently removed. In so doing the hierarchy of grids is refined until only one mesh point is relaxed, which is done exactly. The calculated error is then transferred back down the hierachy of grids as correction term to the field solution. This so called V-cycle is performed several times until a threshold value for the remaining residuum is reached.

Boundary Conditions

Solving the Poisson equation subject to Dirichlet boundary conditions requires the knowledge of the surface potential Φ^s. For an arbitrary distribution of N charges in the system, the evaluation on each boundary grid point j consists of the sum $\Phi_j^{(s)} = \sum_{i=1}^{N} q_i G(|\mathbf{r}_{s,j} - \mathbf{r}_i|)$, where G is the Green's function. Assuming that the number of grid points increases linearly with number of particles, this approach results in a complexity of $\mathcal{O}(N^{5/3})$. This would mean that the determination of the boundary potential would be more time consuming from a certain number of particles than the evaluation of the global field $\hat{\Phi}$ by multigrid methods. To avoid this drawback, a multipole expansion method is used here, which scales linearly in the number of particles. The surface potential is thereby evaluated as

$$\Phi^{(s)}(\mathbf{r}_s) = \sum_{i=1}^{N} \frac{q_i}{|\mathbf{r}_s - \mathbf{r}_i|} \tag{4}$$

$$= \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \omega_l^m(\{\mathbf{r}\}) M_l^m(\mathbf{r}_s) \tag{5}$$

The sum over all charges q_i is therefore split into two terms which factorise into two functions depending only on the set of particle coordinates $\{\mathbf{r}\}$ and the surface coordinates. Therefore the function ω_l^m is only calculated once for a given particle distribution as

$$\omega_l^m(\{\mathbf{r}\}) = \sum_{i=1}^{N} q_i O_l^m(\mathbf{r}_i) \tag{6}$$

and used as a prefactor for the boundary potential of each grid point $\mathbf{r}_s$. The multipole expansions $M_l^m(\mathbf{r}_s)$ and $O_l^m(\mathbf{r}_i)$ are given by [8]

$$O_l^m(\mathbf{r}_i) = r_i^l \frac{1}{(l+|m|)!}\, P_l^m(\cos(\vartheta_{r_i}))e^{-im(\varphi_{r_i})} \tag{7}$$

$$M_l^m(\mathbf{r}_s) = \frac{1}{r_s^{l+1}}(l-|m|)!\, P_l^m(\cos(\vartheta_{r_s}))e^{im(\varphi_{r_s})} \tag{8}$$

and the P_l^m are associated Legendre polynomials. The angles ϑ_x and φ_x are spherical coordinates of the positions $\mathbf{r}_i$ and $\mathbf{r}_s$ in the given coordinate system. Validity of the multipole expansion is only given, if $r_i/r_s < 1$. Therefore the length of the computational domain L_B has to be larger than the region of length L, where charges are located, i.e. $L_B \geq \sqrt{3}L$. For practical purposes, it is chosen here $L_B = 2L$.

Self Energy and Near Field Correction

The solution of the discretized Poisson equation gives a global potential surface $\hat{\Phi}_h(\{\mathbf{r}\})$, including contributions from *all* charges. To calculate the interaction energy $u_i = q_i\phi_i$ of particle i with all other particles, one has to find the proper potential function $\hat{\phi}_{i,h} = \hat{\Phi}_h(\{\mathbf{r}\}|\mathbf{r}_i \notin \{\mathbf{r}\})$, i.e. the potential which would be obtained if particle i was not considered in the solution of Poisson's equation. The solution of this problem is to subtract the self energy contribution from $\hat{\Phi}(\{\mathbf{r}\})$. It is clear that the superposition principle is valid in electrostatics. Therefore the global solution of $\hat{\Phi}_h$ would also be obtained if a sum over all single particle potentials $\hat{\phi}_{i,h}$ would be performed, i.e.

$$\hat{\Phi}_h(\{\mathbf{r}\}) = \sum_{h'} q_{h'}\hat{G}(|\mathbf{r}_h - \mathbf{r}_{h'}|) \tag{9}$$

where $\hat{G}(|\mathbf{r}_h - \mathbf{r}_{h'}|)$ is the grid based Green's function, which is finite for $|\mathbf{r}_h - \mathbf{r}_{h'}| = 0$. This function depends on the solver, which is used in the multigrid cycle. Conveniently it is calculated numerically, placing a unit charge in the centre of a grid with mesh size h and solving Poisson's equation for this problem. In general, particles are located between grid points and their charge is smeared onto the grid. In a 3-dimensial system this results in eight partial charges $\hat{q}_{i,h}$ for each particle. Therefore each particle gets contributions from eight neighbor grid points to its potential and all these potential values $\hat{\phi}_{i,h}$ have to be corrected.

Due to the grid based solution there will be discritization errors, which are most pronounced in the neighbor regions of a particle. Therefore, a more accurate solution will be obtained when applying also near field corrections, i.e. subtracting the grid based solution and calculate explicitly pair-interactions from the neighbored grid cells. Therefore the whole potential energy for particle i is given as

$$\phi_i = \sum_{h \in C_i} w(\mathbf{r}_i - \mathbf{r}_h)\hat{\phi}_{i,h}^{(far)} + \phi_i^{(near)} \tag{10}$$

where

$$\hat{\phi}_{i,h}^{(far)} = \hat{\Phi}_h(\mathbf{r}_i) - \hat{\phi}_{i,h}^{(self)} - \hat{\phi}_{i,h}^{(near)} \quad (11)$$

$$\phi_i^{(near)} = \sum_{j \in C_i \oplus C_n} \frac{q_j}{|\mathbf{r}_i - \mathbf{r}_j|} \quad (12)$$

where C_i is the cell, where particle i is located and $w(\mathbf{r}) = w(x)w(y)w(z)$ is the CIC weighting function (cf. Eq.3). Furthermore, the grid based self energy and near field energy contributions are given by

$$\hat{\phi}_{i,h}^{(self)} = \sum_{h' \in C_i} \hat{q}_{i,h'} \hat{G}(|\mathbf{r}_h - \mathbf{r}_{h'}|) \quad (13)$$

$$\hat{\phi}_{i,h}^{(near)} = \sum_{h' \in C_i \oplus C_n} \sum_{\substack{j \in C_i \oplus C_n \\ j \neq i}} \hat{q}_{j,h'} \hat{G}(|\mathbf{r}_h - \mathbf{r}_{h'}|) \quad (14)$$

In Eq.13 the sum extends over all grid points of cell C_i, where particle i is located. In Eqs.12,14 $C_i \oplus C_n$ is the local cell plus neighbor cells C_n which are taken into account for the near field correction. The first sum in Eq.14 runs over all grid points, belonging to cells, the second over all particles, located in these cells.

Evaluation of Forces

The next step in using the described method for molecular simulations is to calculate the forces. To this end the grid based electric fields are calculated via a finite difference scheme of the potential differences, while the near field part is calculated explicitly, i.e.

$$\hat{E}_{i,h,\alpha}^{(far)} = -\frac{1}{h_\alpha}\left(\hat{\phi}_{i,h+1} - \hat{\phi}_{i,h-1}\right) \quad (15)$$

$$E_{i,\alpha}^{(near)}(\mathbf{r}_i) = \sum_{\substack{j \in C_i \oplus C_n \\ j \neq i}} q_j \frac{(r_{i,\alpha} - r_{j,\alpha})}{|\mathbf{r}_i - \mathbf{r}_j|^3} \quad (16)$$

where $h \pm 1$ means grid point h plus or minus 1 in direction α. The resulting forces are then obtained from an interpolation of grid functions to particle i, i.e.

$$F_{i,\alpha} = q_i \sum_{h \in C_i} w(\mathbf{r}_i - \mathbf{r}_h) \hat{E}_{i,h,\alpha}^{(far)} + q_i E_{i,\alpha}^{(near)} \quad (17)$$

3 Results and Conclusions

In order to test the performance of the method, systems with different numbers of particles were calculated with the multigrid method and compared with explicit pair wise summations. For the test cases equilibrated systems of Lennard-Jones particles were chosen. For electrostatic test calculations, charges of ± 1

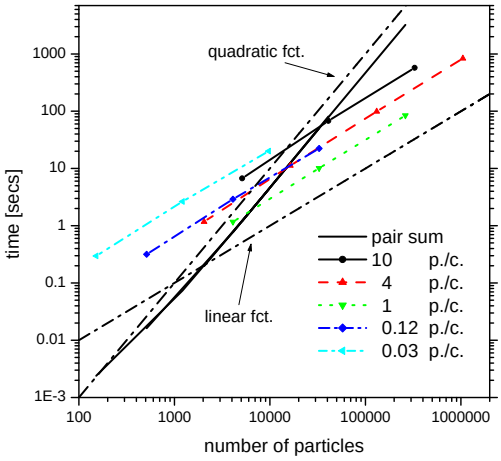

Figure 1: Timings for multigrid method compared with explicit pairwise summation for different average number of particles per cell.

were distributed randomly onto particles subject to the condition $\sum_{i=1}^{N} q_i = 0$. The boundary potential was calculated with a multipole expansion up to level $l = 10$. Fig.1 shows results for different cases, where the average number of particles per cell was varied. According to the number of particles, the number of grid points n_h in each direction was varied between $n_h = 2^3 + 1, \ldots, 2^6 + 1$. First of all the linear increase of CPU time with number of particles is recovered from the figure. Furthermore it is seen that one particle per cell is the fastest version. A crossover at ≈ 5000 particles is observed, where the multigrid algorithm gets faster than explicit summations. It is furthermore found that relative error norms, $\|\mathbf{F} - \mathbf{F}^{(ex)}\|/\|\mathbf{F}^{(ex)}\|$ of the forces are in the range of $\approx 2\%$. This estimate refers to the choice of one neighbor cell layer for the near field part, i.e. $\#(C_n) = 3^3 - 1 = 26$ in Eqs. 12,14. Taking into account more layers of neighbor cells ($\#(C_n) = 5^3 - 1, 7^3 - 1, \ldots$), will increase the accuracy, but will also decrease the performance. On the lowest level, this makes the method at the moment only moderatley accurate, compared with other methods. Higher accuracy is to be expected by using a more refined splitting of near and far field contributions, by increasing the near field part (as mentioned), by other methods of charge assignment to the mesh points or higher order approximations to Eq. 15. Finally we note that the method will also be applicable for periodic boundary conditions. In this case no multipole expansion of the boundary potential is required and slightly better results are to be expected. Work in this direction is in progress.

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