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LINEAR SCALING SOLUTION OF THE COULOMB PROBLEM USING WAVELETS

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The Coulomb problem for continuous charge distributions is a central problem in physics. Powerful methods, that scale linearly with system size and that allow us to use different resolutions in different regions of space are therefore highly desirable. Using wavelet based Multi Resolution Analysis we derive a novel method which has these properties. The power and accuracy of the method is illustrated by applying it to the calculation of the electrostatic potential of a full three-dimensional all-electron uranium dimer. © 1998 Elsevier Science Ltd

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The theory of wavelets [1, 2] is one of the most important recent development in mathematics. It allows one to apply a multi-scale analysis to problems that exhibit widely varying length scales [1]. Problems with this feature abound in all fields of physics. The problem we want to address here is the classical Coulomb problem for a continuous charge distribution ρ, i.e. we want to solve Poisson's equation

$$\nabla^2 V = -4\pi\rho \tag{1}$$

under the constraint that the potential V vanishes at infinity. This basic equation can be found in nearly any field of physics and it is therefore essential to have efficient solution methods for it. There are two important requirements for an algorithm that solves this problem. First it should scale linearly with the "size" of the charge distribution. By size we mean the number of basis functions necessary to represent the charge distribution. This linear scaling property is of utmost importance if one calculates very large systems. Second, it should allow for varying resolutions, so that one can increase the resolution in regions where this is required. For discrete charge distributions several algorithms [6] with these two properties exist and have become a standard for simulations of large coulombic and gravitational particle systems. For continuous charge distributions proposals have been put forward to map the continuous problem onto a discrete and to use the above mentioned algorithms for discrete systems [7]. To the best of our knowledge, there exists however no linear scaling algorithm for nonuniform grids that can directly be applied to continuous charge distributions. If one constrains oneself to uniform grids and periodic boundary conditions, there are of course the well known Fourier techniques [8], that show a nearly linear $N \log_2(N)$ scaling with respect to the number of grid points N. Non-periodic boundary conditions can be implemented in the context of Fourier techniques only by cutting off the long range Coulomb potential [4]. Finite element methods allow nonuniform grids, but grid generation and preconditioning pose severe problems. Multigrid methods [5] share several features with the method to be presented, but no calculations of physical systems exhibiting widely different length scales have been presented so far. Using a basis of wavelet functions, we will present in this paper a method that scales strictly linear and allows for nonuniform grids.

There are many families of wavelets and one has to choose the most appropriate one for a specific application. A widely used family are the compactly supported orthogonal wavelets of Daubechies [2]. The orthogonality property is convenient if one has to expand an arbitrary function in a basis of wavelets. Their disadvantage is that a high degree of smoothness (i.e. many continous derivatives) can only be obtained at the cost of a rather long support length which would substantially slow down all the necessary

computations. For a given support length, one can however construct families of nonorthogonal wavelets that are much smoother. The so-called coiflets [2] as well as the non-orthogonal wavelets constructed by Daubechies [2] and the so-called second generation wavelets of Sweldens [10] fall into this category. In principle they would all be suitable as a basis for the method we are going to present. The reason why we decided to use second generation interpolating wavelets [9, 10] is that for this class of wavelets the mapping [3] from the numerical values on a grid to the expansion coefficients of the wavelet basis is easiest and that they give rise to a particularly fast wavelet transform [10].

Wavelets have already been successfully applied in several areas of physics [11]. In the context of electronic structure calculations a fairly small wavelet basis can describe the widely varying scales of both core and valence electrons [12]. Self-consistent electronic structure calculations have also been done [13]. In these self-consistent calculations the solution of Poisson's equation was however done by traditional Fourier techniques.

In addition to the scaling function ϕ and the wavelet ψ of ordinary wavelet theory biorthogonal wavelet theory [2] still has the complementary dual scaling function $\tilde{\phi}$ and the dual wavelet $\tilde{\psi}$. Each scaling function and wavelet belongs to a hierarchical level of resolution. By analysing a function with respect to these different levels of resolution one can do a so-called Multi Resolution Analysis (MRA). The space belonging to a certain level of resolution k is spanned by all the integer translations of the scaling function $\phi_{i,k}(x) \propto \phi((\frac{1}{2})^k x - i)$. Introducing a projection operator P_k that projects any function f onto the space of resolution k we can write

$$P_k[f(x)] = \sum_i s_{i,k}\phi_{i,k}(x) . \qquad (2)$$

Since the scaling functions and their complementary counterparts are orthogonal, ($< \phi_{k,i}(x) \mid \tilde{\phi}_{k,j}(x) > = \delta_{ij}$), the expansion coefficients $s_{i,k}$ are given by

$$s_{i,k} = < f(x) \mid \tilde{\phi}_{i,k}(x) > . \qquad (3)$$

The projected function $P_k[f(x)]$ approches the original function f if one decreases k and it becomes identical in the limit $k \to -\infty$. This is an important feature because it allows us to improve systematically the numerical accuracy in very much the same way as it is done with a basis of plane waves. In numerical application it is of course not possible to take this limit, and we will therefore denote the finest level of resolution that is used in the calculation by $k = 0$

The scaling function satisfies a refinement relation

$$\phi_{j,k}(x) = \sum_l \tilde{h}_{l-2j}\phi_{l,k-1}(x)$$

i.e. each scaling function of a lower resolution level can be expressed as a linear combination of higher resolution scaling functions. It is obviously not possible to express a scaling function of higher resolution by a linear combination of lower resolution scaling functions only. One can, however, write down such an expression if one still includes the wavelets $\psi_{i,k}(x) \propto \psi((\frac{1}{2})^k x - i)$

$$\phi_{j,k-1}(x) = \sum_l h_{l-2j}\phi_{l,k}(x) + \sum_l g_{l-2j}\psi_{l,k}(x) . \qquad (4)$$

The wavelets at level k thus reintroduces the resolution that is lost as one goes from level k to level $k+1$ scaling functions. One can introduce a projection operator Q_k that project any function onto the wavelet space at resolution k and in analogy to equation (2), 3 one obtains

$$Q_k[f(x)] = \sum_i d_{i,k}\psi_{i,k}(x) \; ; \; d_{i,k} = < f(x) \mid \tilde{\psi}_{i,k}(x) > .$$

The scaling function and wavelet expansion coefficients at the different level are related by

$$s_{i,k+1} = \sum_j h_{i-2j}s_{j,k} \; ; \; d_{i,k+1} = \sum_j g_{i-2j}s_{j,k} \qquad (5)$$

$$s_{i,k} = \sum_j \left(\tilde{h}_{i-2j}s_{i,k+1} + \tilde{g}_{i-2j}d_{i,k+1}\right) . \qquad (6)$$

Equation (5) is called a forward fast wavelet transform, equation (6) being its inverse counterpart. If one has periodic boundary conditions for the data $s_{i,k}$ the wavelet transform is one-to-one transformation between $s_{i,k}$ and its spectral decompositions $s_{i,k+1}$ and $d_{i,k+1}$. To obtain a full wavelet spectral analysis the forward transform is applied recursively. In consecutive steps the output data $s_{i,k+1}$ of the previous forward transform are the input data for the next transform. The size of the data set to be transformed is thus cut into half in each step. The total operation count is then given by a geometric series and scales therefore strictly linear. A full wavelet synthesis consists in the same way of a sequence of inverse transforms and gives back the original data set $s_{i,k}$. The coefficients h_i and g_i and their complementary counterparts $\tilde{h}_i$, $\tilde{g}_i$ are filters of finite length 2 m and can be derived from the MRA requirements [2]. The 8-th order lifted Lazy scaling function and wavelet [10] that were used in this work are shown in Fig. 1. Because it can represent polynomials up to degree 8 exactly, the expansion coefficients with respect to the wavelets $d_{i,k}$ decay very rapidly for any smooth function with decreasing k.

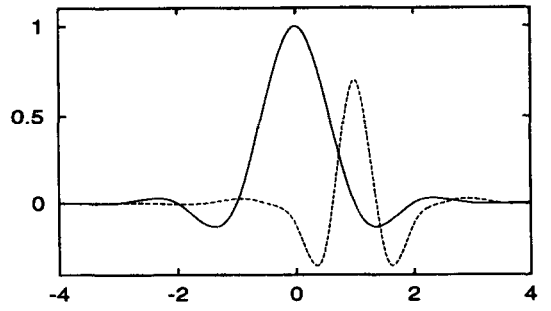

Fig. 1. The scaling function (full line) and wavelet (dashed line) used in this work.

To do a multidimensional MRA, we use a scheme described by Daubechies [2]. Even though all this work was done in the three-dimensional case, we will illustrate the principle just for the two-dimensional case. The space of all scaling functions of resolution level k is given by

$$\phi_{i,j,k}(x, y) = \phi_{i,k}(x)\phi_{j,k}(y). \tag{7}$$

Using equation (4) one obtains three kind of terms for the wavelet space

$$\psi^{01}_{i,j,k}(x, y) = \phi_{i,k}(x)\psi_{j,k}(y) \tag{8}$$

$$\psi^{10}_{i,j,k}(x, y) = \psi_{i,k}(x)\phi_{j,k}(y) \tag{9}$$

$$\psi^{11}_{i,j,k}(x, y) = \psi_{i,k}(x)\psi_{j,k}(y). \tag{10}$$

Given the expansion coefficients of the potential $V_{i,j,k}$ and of the charge density $\rho_{i,j,k}$, Poisson's equation leads to the system of equations

$$\sum_{j1,j2} L_{i,j;\mu,\nu} V_{\mu,\nu} = \rho_{i,j}$$

where

$$L^k_{i,j;\mu,\nu} = < \tilde{\phi}_{i,k}(x)\, \tilde{\phi}_{j,k}(y) | \nabla^2 | \phi_{\mu,k}(x)\, \phi_{\nu,k}(y) > .$$

Since the scaling functions have a finite support, the matrix L^k is a sparse matrix and its nonzero elements $L^k_{i1,i2;j1,j2}$ can be calculated analytically [14]. The natural boundary conditions for this scheme are periodic boundary conditions. As we stressed in the introduction, we however want to solve Poisson's equation (1) with non-periodic boundary conditions. Since boundary effects vanish whenever the boundary is sufficiently far away, one could in principle obtain natural boundary conditions (i.e. $V(r) \to 0$ if $r \to \infty$) within arbitrary precision by using a sufficiently large periodic box. Since the electrostatic potential decays fairly slowly a very large box would be required and the numerical effort would be tremendous if one uses equally spaced grids within this huge periodic computational box. Far away from the charge distribution

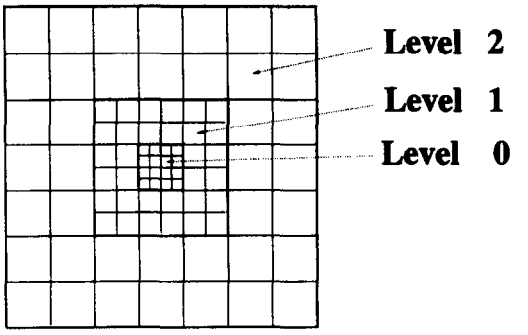

Fig. 2. Regions of different resolution as obtained with the wavelet scheme. For simplicity only three levels of resolution are shown.

the variation of the potential is however small and less resolution is needed. The key idea is therefore to use a basis of scaling functions at the highest (periodic) level and then to increase the resolution towards the center by adding there wavelets of higher resolution. They resulting regions of different resolutions are shown in Fig. 2. Note that within this 3-dimensional product scheme all the refinement region have to be rectangles.

Up to now the motivation for introducing grids of different resolution was to handle the natural boundary conditions. Additional levels of resolution can however be introduced to handle charge distributions that have different length scales and require therefore higher resolution in some parts of space. The theory of wavelets gives us also enough flexibility to increase the resolution not only around one center but around any number of centers in the computational box.

A full wavelet synthesis step can be done straightforwardly in this hierarchical setting. Any wavelet can be decomposed into scaling functions and therefore one can calculate the scaling function coefficients at any level of resolution and for any point in the computational volume. If one calculates these scaling functions for high resolution levels in a region of low resolution, one obtains however a highly redundant data set. To do a full wavelet analysis that brings back the original spectral decomposition data, it turns out that one needs actually a slightly redundant data set. In order to calculate the wavelet coefficients for a wavelet at a boundary to a lower resolution region, one needs the scaling function values corresponding to this higher resolution also in a strip of width m in the lower resolution region. A schematic diagram of a full hierarchical wavelet analysis and synthesis is shown in Fig. 3.

In this mixed representation, where one has scaling functions at the highest periodic level and wavelets all the refinement levels, the structure of the Laplace operator is much more complicated since one has coupling between all the hierarchical levels. An elegant way to cope with this additional complexity is

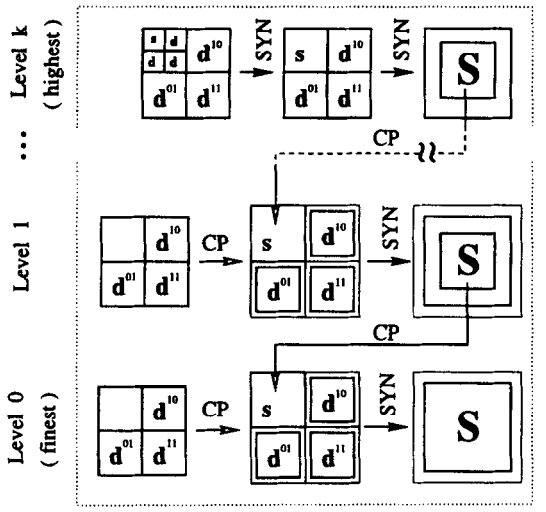

Fig. 3. A schematic representation of a multi hierarchy wavelet synthesis. Data regions denoted by s,d^{01},d^{10} and d^{11} contain expansion coefficients for basis functions of the type given by equations (7), (8), (9) and (10) respectively. CP stand for a copy step where one puts an additional layer of zeroes around the data set. SYN denotes a one level wavelet synthesis step. One starts the process at the coarsest (periodic) level and proceeds down to the finest resolution level. To do a multi hierarchy wavelet analysis one proceeds back up reversing all the copy operations and replacing the single level synthesis steps by analysis steps.

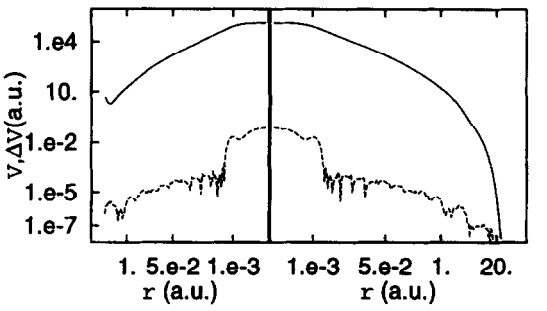

Fig. 4. The potential V (full line) and the numerical error ΔV (dashed line) for an uranium dimer as a function of the distance from of right hand side nucleus. The left panel shows both quantities in the direction of the left nucleus up to the middle of the bond, the right panel shows them in the opposite direction. Both distances are given on a logarithmic scale. The behaviour of both quantities in the other regions of space is very similar.

the so-called nonstandard operator form proposed by Beylkin, Coifman and Rokhlin [15], which allows us to incorporate this coupling by a sequence of wavelet transforms (Fig. 3), that are interleaved with the application of a simple one-level Laplace operator. For this one-level Laplace operator only the matrix elements of the Laplace operator among scaling functions and wavelets of the same resolution level, but not between different levels of resolution are needed. This technique can also be used for the evaluation of the Laplace operator in the context of electronic structure calculations [16].

Mathematically the nonstandard operator form is a telescopic expansion of the Laplace operator in the scaling function basis at the finest level L^0. The projection operators P_k and Q_k, and their dual counterparts $\tilde{P}_k$ and $\tilde{Q}_k$ satisfy

$$P_k = P_{k+1} + Q_{k+1} \quad ; \quad \tilde{P}_k = \tilde{P}_{k+1} + \tilde{Q}_{k+1}$$

and we may write

$$L^k = \tilde{P}_k L^0 P_k = (\tilde{Q}_{k+1} + \tilde{P}_{k+1}) L^0 (Q_{k+1} + P_{k+1}) = L_{DD}^{k+1} + L_{SD}^{k+1} + L_{DS}^{k+1} + L^{k+1} \quad (11)$$

where L_{DD}^{k+1}, L_{SD}^{k+1}, L_{DS}^{k+1} are Laplace operators at the $(k+1)$th level representing the coupling of wavelets with wavelets, wavelets with scaling functions and scaling functions with wavelets. Applying equation (11) recursively for $k = 0, 1, \ldots$, one obtains the nonstandard operator form. Let us note that the nonstandard form uses redundant data sets where both the scaling functions and wavelet coefficients are needed.

In this basis of lifted interpolating wavelets at different resolution levels a simple diagonal preconditioning scheme is very efficient and independently of the number of resolution levels we were able to reduce the residue by one order of magnitude with only 3 iterations.

To demonstrate the power of this method we applied it to a problem that can hardly be solved by any other method, namely the potential arising from the nucleonic and electronic charge distribution of a fully three-dimensional all-electron uranium dimer. The charge distribution of the nucleus was represented by a Gaussian charge distribution with an extension of $\frac{1}{2000}$ atomic units. Since the valence electrons have an extension which is of the order of one atomic unit, we have length scales that differ by more than 3 orders of magnitude. As can be seen from Fig. 4, the potential also varies by many orders of magnitude. Using 22 hierarchical levels in our algorithm, we can represent resolutions that differ by 7 orders of magnitude and we are able to calculate the potential with at least 6 significant digits in the whole region from the nucleus to the valence region. In order to be able to determine the error we actually first fitted the electronic charge distribution by a small number of Gaussians, whose exact potential can be calculated analytically. This rather crude charge density was then used in all the calculations.

We also applied the method to clusters containing

pseudo-potential *CO* molecules. In this case there is only one length scale associated with the charge distribution and it is possible to reduce the number of grid points on higher levels such that the total amount of work increases only slightly with additional hierarchies. We were able to calculate the potential corresponding to the non-periodic boundary conditions with 8 significant digits.

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