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Grid-based lattice summation of electrostatic potentials by assembled rank-structured tensor approximation

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ABSTRACT

Our recent method for low-rank tensor representation of sums of the arbitrarily positioned electrostatic potentials discretized on a 3D Cartesian grid reduces the 3D tensor summation to operations involving only 1D vectors however retaining the linear complexity scaling in the number of potentials. Here, we introduce and study a novel tensor approach for fast and accurate assembled summation of a large number of lattice-allocated potentials represented on 3D $N \times N \times N$ grid with the computational requirements only *weakly dependent* on the number of summed potentials. It is based on the assembled low-rank canonical tensor representations of the collected potentials using pointwise sums of shifted canonical vectors representing the single generating function, say the Newton kernel. For a sum of electrostatic potentials over $L \times L \times L$ lattice embedded in a box the required storage scales linearly in the 1D grid-size, $O(N)$, while the numerical cost is estimated by $O(NL)$. For periodic boundary conditions, the storage demand remains proportional to the 1D grid-size of a unit cell, $n = N/L$, while the numerical cost reduces to $O(N)$, that outperforms the FFT-based Ewald-type summation algorithms of complexity $O(N^3 \log N)$. The complexity in the grid parameter N can be reduced even to the logarithmic scale $O(\log N)$ by using data-sparse representation of canonical N-vectors via the quantics tensor approximation. For justification, we prove an upper bound on the quantics ranks for the canonical vectors in the overall lattice sum. The presented approach is beneficial in applications which require further functional calculus with the lattice potential, say, scalar product with a function, integration or differentiation, which can be performed easily in tensor arithmetics on large 3D grids with 1D cost. Numerical tests illustrate the performance of the tensor summation method and confirm the estimated bounds on the tensor ranks.

1. Introduction

There are several challenges in the numerical treatment of periodic and perturbed periodic systems in quantum chemical computations for crystalline, metallic and polymer-type compounds, see [1–8]. One of them is the lattice summation of electrostatic potentials of a large number of nuclei distributed on a fine 3D computational grid. This problem is also considered to be a demanding computational task in the numerical treatment of long-range electrostatic interactions in molecular dynamics simulations of large solvated biological systems [9–11]. In the latter applications the efficient calculation of quantities like potential energy function or interparticle forces remains to be of main interest.

Tracing back to Ewald summation techniques [12], the development of lattice-sum methods in numerical simulation of particle interactions in large molecular systems has led to established algorithms for evaluating long-range electrostatic potentials of multiparticle systems, see for example [13,14,9–11,15] and references therein. These methods usually combine the original Ewald summation approach with the Fast Fourier Transform (FFT) or fast multipole methods [16].

The Ewald summation techniques were shown to be particularly attractive for computation of the potential energies and forces of many-particle systems with long-range interaction potential in periodic boundary conditions. They are based on the spacial separation of a sum of potentials into two parts, the short-range part treated in the real space, and the long-range part whose sum converges in the reciprocal space. The fast multipole method is used for more unstructuredly distributed potentials, where the interactions between closely positioned potentials are calculated directly, and the distant interactions are calculated by using the hierarchical clusters.

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Here we propose the new approach for calculation of lattice sums based on the assembled low-rank tensor-product approximation of the electrostatic potentials (shifted Newton kernels) discretized on large $N \times N \times N$ 3D Cartesian grid. A sum of potentials is represented on the 3D uniform grid in the whole computational box, as a low-rank tensor with storage $O(N)$, containing tensor-products of vectors of a special form. This remarkable approach is initiated by our former numerical observations in [17,18] that the Tucker tensor rank of the 3D lattice sum of discretized Slater functions remains uniformly bounded, nearly independent of the number of single Slater functions in a sum.

As a building block we use the separable tensor-product representation (approximation) of a single Newton kernel $\frac{1}{r}$ in a given computational box, which provides the electrostatic potential at any point of an $N \times N \times N$-grid, but needs only $O(N)$ storage due to the canonical tensor format (A.1). This algorithm, introduced in [19], includes presentation of the potential in a form of a weighted sum of Gaussians obtained by the sinc-quadrature approximation to the integral Laplace transform of the kernel function $\frac{1}{r}$, [20,21]. Then by shifting and summation of the single canonical tensors one can construct a sum of electrostatic potentials located at the positions of nuclei in a molecule. This scheme of *direct* tensor summation was introduced in [22,23] for the calculation of the one-electron integrals in the black-box Hartree–Fock solver by grid-based tensor numerical methods.[1] It is well suited for the case of arbitrary positions of potentials, like for example nuclei in a molecule, however the rank of the resulting canonical tensor is approximately proportional to a number of summed potentials.

In this paper we introduce a novel grid-based *assembled* tensor summation method which matches well for lattice-type and periodic molecular systems and yields enormous reduction in storage and time of calculations. The resulting canonical tensor representing the total sum of a large number of potentials contains the same number of canonical vectors as a tensor for a single potential. However, these vectors have another content: they collect the whole data from the 3D lattice by capturing the periodic shape of the total 3D potential sum (represented on the grid) onto a few 1D canonican vectors, as it is shown in Figs. 3.2 and 3.5 in sections Sections 3.1 and 3.2. This agglomeration is performed in a simple algebraic way by pointwise sums of shifted canonical vectors representing the generating function, e.g. $\frac{1}{r}$. The presented numerical calculations confirm that the difference between the total potentials obtained by an assembled tensor-product sum and by a direct canonical sum is close to machine precision, see Fig. 3.4.

Thus, the adaptive global decomposition of a sum of interacting potentials can be computed with a high accuracy, and in a completely algebraic way. The resultant potential is represented simultaneously on the fine 3D Cartesian grid in the whole computational box, both in the framework of a finite lattice-type cluster, or of a supercell in a periodic setting. The corresponding rank bounds for the tensor representation of the sums of potentials are proven. Our grid-based tensor approach is beneficial in applications requiring further functional calculus with the lattice potential sums, for example, interpolation, scalar product with a function, integration or differentiation (computation of energies or forces), which can be performed on large 3D grids using tensor arithmetics of sub-linear cost [18,24] (see Appendix). This advantage makes the tensor method promising in electronic structure calculations, for example, in computation of projections of the sum of electrostatic potentials onto some basis sets like molecular or atomic Gaussian-type orbitals.

[1] The accuracy of tensor-based calculations is close to accuracy of benchmark Hartree–Fock packages based on analytical evaluation of the corresponding integrals [22,23].

In the case of an $L \times L \times L$ lattice cluster in a box the storage size is shown to be bounded by $O(L)$, while the summation cost is estimated by $O(NL)$. The latter can be reduced to the logarithmic scaling in the grid size, $O(L \log N)$, by using the quantized approximation of long canonical vectors (QTT approximation method [25], see Appendix). For a lattice cluster in a box both the fast multipole, FFT as well as the so-called P^3M methods if applicable scale at least linear-logarithmic in the number of particles/nuclear charges on a lattice, $O(L^3 \log L)$, see [14,11].

For periodic boundary conditions, the respective lattice summations are reduced to 1D operations on short canonical vectors of size $n = N/L$, being the restriction (projection) of the global N-vectors onto the unit cell. Here n denotes merely the number of grid points per unit cell. In this case, storage and computational costs are reduced to $O(n)$ and $O(Ln)$, respectively, while the traditional FFT-based approach scales at least cubically in L, $O(L^3 \log L)$. Due to low cost of the tensor method in the limit of large lattice size L, the conditionally convergent sums in periodic setting can be regularized by subtraction of the constant term which can be evaluated numerically by the Richardson extrapolation on a sequence of lattice parameters L, $2L$, $4L$ etc. (see Section 3.2). Hence, in the new framework the analytic treatment of the conditionally convergent sums is no longer required.

It is worth to note that the presented tensor method is applicable to the lattice sums of rather general interaction potentials which allow an efficient local-plus-separable approximation. In particular, along with Coulombic systems, it can be applied to a wide class of commonly used interaction potentials, for example, to the Slater, Yukawa, Stokeslet, Lennard-Jones or van der Waals interactions. In all these cases the existence of low-rank grid-based tensor approximation can be proved and this approximation can be constructed numerically by analytic–algebraic methods as in the case of the Newton kernel. Our tensor approach can be extended to slightly perturbed periodic systems, for example, to the case of few vacancies in the spacial distribution of electrostatic potentials, or a small perturbation in positions of electron charges and other defects. The more detailed discussion of these issues is beyond the scope of the present paper, and is the topic of forthcoming papers.

Notice that the tensor numerical methods are now recognized as a powerful tool for solution of multidimensional partial differential equations (PDEs) discretized by traditional grid-based schemes. Originating from the DMRG-based matrix product states decomposition in quantum physics and chemistry [26] and coupled with tensor multilinear algebra [27–29], the approach was recently developed to the new branch of numerical analysis, tensor numerical methods, providing efficient algorithms for solving multidimensional PDEs with linear complexity scaling in the dimension [30]. One of the first steps in the development of tensor numerical methods was the 3D grid-based tensor-structured method for solution of the nonlinear Hartree–Fock equation [31,32,18,23] based on the efficient algorithms for the grid-based calculation of the 3D convolution integral operators in 1D complexity.

The remainder of the paper is structured as follows. In Section 2 we recall the low-rank approximation to the single Newton kernel (electrostatic potential) in the canonical tensor format and direct tensor calculation of the total potential sum of arbitrarily positioned potentials in a box. Section 3 presents the main results of this paper describing the assembled low-rank tensor summation of potentials on a lattice in a bounded rectangular box, as well as in the periodic setting. The storage estimates and complexity analysis are provided. We give numerical illustrations to the structure of assembled canonical vectors and the results on accuracy and times of tensor summations over large 3D lattice. In Section 4, we prove the low QTT-rank approximation of the canonical vectors in the lattice sum of the Newton kernels that justifies the logarithmic