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MRA and low-separation rank approximation with applications to quantum electronics structures computations

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Abstract. We describe some recent mathematical results in constructing computational methods that lead to the development of fast and accurate multiresolution numerical methods for solving problems in computational chemistry (the so-called multiresolution quantum chemistry).

Using low separation rank representations of functions and operators and representations in multiwavelet bases, we developed a multiscale solution method for integral and differential equations and integral transforms. The Poisson equation and the Schrodinger equation, the projector on the divergence free functions, provide important examples with a wide range of applications in computational chemistry, computational electromagnetic and fluid dynamics.

We have implemented these ideas that include adaptive representations of operators and functions in the multiwavelet basis and low separation rank approximation of operators and functions. These methods have been implemented into a software package called Multiresolution Adaptive Numerical Evaluation for Scientific Simulation (MADNESS).

1. Introduction

It was already clear in [3, 4] that multiresolution representations of functions and operators should lead to useful numerical algorithms. However, the straightforward generalization of such approach from one spatial dimension to that in higher dimensions yields algorithms that are too costly for practical applications. The development of efficient and robust algorithms using multiresolution analysis (MRA) for solving partial differential and integral equations in three and higher dimensions has only recently been successful [5, 12, 13, 14]. These algorithms rely on low separation rank (LSR) representation [6, 7] and tools for constructing a particular class of such representations [8, 14] using approximations via Gaussian.

Approximations involving Gaussian have a long history in quantum chemistry, starting with [9, 17, 16]. Typically Gaussian (usually with an additional polynomial factor) approximate the wave functions, including their cusps. More recently, an approximation by Gaussian of the function $1/r$ (viewed as a nuclear potential) has been considered in [15, 10].

In our approach we use Gaussians to construct separated representations of Green's functions as the first step in obtaining their efficient multiresolution representation. In [5, 12, 13, 14] we developed a MRA approach (using multiwavelet bases) which incorporates the advantages of convenient adaptive refinement in representing functions and operators, guaranteed solution for arbitrary (finite) precision and that of a fast algorithm. The speed of our algorithms is comparable to that of the Fast Multipole Method (FMM) [11]. In fact, the approximation technique used in our approach and that in FMM are related but this topic is outside the scope of this paper. We only note that our approach has advantages in higher dimensions.

We also note that the approach in [8, 14] is applicable to a wide variety of kernels, including singular kernels such as the projector on the divergence free functions and some oscillatory kernels, e.g., the Helmholtz kernel $\frac{e^{-ikr}}{r}$, where kr is moderately large in the region of interest. As a result, our approach opens up new opportunities in constructing effective numerical methods and points out a way of developing practical operator calculus in high dimensions. In particular, we are working on constructing multiparticle Green's functions and spectral projectors. In this paper we briefly summarize some of our work and that of our collaborators in systematically applying MRA and LSR approximations to problems in computational quantum chemistry. We also outline current research and new directions that originate in our approach.

2. Low Separation Rank Representation

The separated representation of a multivariable function f is an approximation

$$\|f(x_1, x_2, \dots, x_n) - \sum_{l=1}^r s_l \varphi_1^l(x_1) \varphi_2^l(x_2) \dots \varphi_n^l(x_n)\| \leq \epsilon. \quad (1)$$

We set the desired accuracy ϵ first, and then adapt the functions $\{\varphi_i^l(x_i)\}$ and the numbers $\{s_l\}$ so to achieve such accuracy with a minimal separation rank r . In the same manner, we approximate multidimensional operators as a sum of products of operators acting in each direction separately. The set of functions $\{\varphi_i^l(x_i)\}$ in (1) is not fixed and depends on the function f . The functions $\{\varphi_i^l(x_i)\}$ come from a family of functions that is typically too wide to form a basis (see examples below). Such approach goes beyond the notion of a basis for representing functions; we note that the algorithms for constructing and maintaining (1) are necessarily nonlinear (see [6, 7, 8]).

As an example, let us consider approximating the function $1/r^\alpha$ with a collection of Gaussians. It is shown in [8] (see also [15, 10]) that for any $\alpha > 0$, $0 < \delta \leq 1$, and $0 < \epsilon \leq \min\{\frac{1}{2}, \frac{8}{\alpha}\}$, there exist positive numbers p_m and w_m such that

$$\left| r^{-\alpha} - \sum_{m=1}^M w_m e^{-p_m r^2} \right| \leq r^{-\alpha} \epsilon, \text{ for all } \delta \leq r \leq 1 \quad (2)$$

with

$$M = \log \epsilon^{-1} [c_0 + c_1 \log \epsilon^{-1} + c_2 \log \delta^{-1}], \quad (3)$$

where c_k are constants that only depend on α .

For fixed power α and accuracy ϵ , we have $M = \mathcal{O}(\log \delta^{-1})$. The actual number of terms needed for this approximation is mercifully small, for example, for $\alpha = 1$, $\epsilon = 10^{-8}$ and $\delta = 10^{-9}$, the number of terms $M = 89$. We note that in this case the family of functions from which the functions $\{\varphi_i^l(x_i)\}$ are selected in (1) are Gaussians, e^{-pr^2} , where $p > 0$. The particular set of exponents and coefficients depends on α , ϵ and δ . Similar approximations are also available now for oscillatory functions, e.g. e^{ikr}/r , where the functions $\{\varphi_i^l(x_i)\}$ in (1) are Gaussians, $e^{\tau r^2}$, and τ are complex-valued.

We will now turn to multiresolution representations.

3. Multiresolution Representations

We have chosen multiwavelets [1] as bases for multiresolution representation of operators in two and three dimensions. This choice properly addresses many (contradictory) requirements on such bases, namely, an accommodation of boundary conditions without a loss of order or conditioning of approximation, an efficient algorithm for point-wise multiplication as well as availability of scale-consistent analogues of forward and backward differentiation (see [2]).

In our approach we start with the separated representation of the operator kernels, e.g. as in (2), and then construct a representation in multiwavelet bases. The necessary estimates are available in [5] and corresponding algorithms were developed in [13, 14, 18, 19].

Let us briefly explain the key elements of our approach using an example of convolution with the Poisson kernel, $K(\mathbf{x}) = \frac{1}{4\pi\|\mathbf{x}\|}$, where $-\Delta K(\mathbf{x}) = \delta(\mathbf{x})$, $x \in \mathbb{R}^3$. In order to represent this operator in multiwavelet bases, we need to compute the integrals

$$t_{ii',jj',kk'}^{n;\mathbf{l},\mathbf{l}'} = \int K(\mathbf{x} - \mathbf{y}) \phi_{i,l_1}^n(x_1)\phi_{i',l'_1}^n(y_1) \phi_{j,l_2}^n(x_2)\phi_{j',l'_2}^n(y_2) \phi_{k,l_3}^n(x_3)\phi_{k',l'_3}^n(y_3) d\mathbf{x}d\mathbf{y}, \quad (4)$$

where $\mathbf{x} = (x_1, x_2, x_3)$, $\mathbf{y} = (y_1, y_2, y_3)$, $\mathbf{l} = (l_1, l_2, l_3)$ and $\mathbf{l}' = (l'_1, l'_2, l'_3)$. The integration in (4) is over the support of the product of one-dimensional scaling functions, $\phi_{i,l}^n(x) = 2^{n/2}\phi_i(2^n x - l)$, where ϕ_i are the normalized Legendre polynomials on the interval $[0, 1]$,

$$\phi_i(x) = \begin{cases} \sqrt{2i+1}P_i(2x-1), & x \in [0, 1] \\ 0, & x \notin [0, 1] \end{cases}, \quad (5)$$

and P_i are the Legendre polynomials on $[-1, 1]$, $i = 0, \dots, m-1$, and m is the order of the basis,

By changing variables of integration and taking advantage of K being a homogeneous convolution kernel, we obtain $t_{ii',jj',kk'}^{n;\mathbf{l},\mathbf{l}'} = 2^{-2n} t_{ii',jj',kk'}^{\mathbf{l}-\mathbf{l}'}$ where

$$t_{ii',jj',kk'}^{\mathbf{l}} = t_{ii',jj',kk'}^{l_1,l_2,l_3} = \int_B K(\mathbf{x} + \mathbf{l}) \Phi_{ii'}(x_1) \Phi_{jj'}(x_2) \Phi_{kk'}(x_3) d\mathbf{x}. \quad (6)$$

and $\Phi_{ii'}(x)$ are the cross-correlation of the scaling functions, $\Phi_{ii'}(x) = \int_{-\infty}^{\infty} \phi_i(x+y)\phi_{i'}(y)dy$. Since the supports of the scaling functions are restricted to $[0, 1]$, the functions $\Phi_{ii'}$ are zero outside the interval $[-1, 1]$ and $B = [-1, 1]^3$. The functions $\Phi_{ii'}$ are polynomials on $[-1, 0]$ and $[0, 1]$ of degree $i + i' + 1$.

We note that the total number of coefficients in (6), m^6 for each \mathbf{l} , is too large for a practical method of applying this kernel. To reduce the cost of using coefficients (6), we construct their separated representation for a finite but arbitrary accuracy ϵ . As a result, the computation of coefficients (6) reduces to the evaluation of a small number of one-dimensional integrals.

We prove in [5] that for any $\epsilon > 0$ and $0 < \delta \leq 1$ the coefficients $t_{ii',jj',kk'}^{\mathbf{l}}$ in (6) have an approximation with a low separation rank,

$$r_{ii',jj',kk'}^{\mathbf{l}} = \frac{1}{4\pi} \sum_{m=1}^M \frac{w_m}{b} F_{ii'}^{m,l_1} F_{jj'}^{m,l_2} F_{kk'}^{m,l_3}, \quad (7)$$

such that if $\max_i |l_i| \geq 2$, then

$$|t_{ii',jj',kk'}^{\mathbf{l}} - r_{ii',jj',kk'}^{\mathbf{l}}| \leq \frac{2\epsilon}{\pi}, \quad (8)$$

and if $\max_i |l_i| \leq 1$, then

$$|t_{ii',jj',kk'}^{\mathbf{l}} - r_{ii',jj',kk'}^{\mathbf{l}}| \leq C\delta^2 + \frac{2\epsilon}{\pi}, \quad (9)$$

where $b = \sqrt{3} + \|\mathbf{l}\|$,

$$F_{ii'}^{m,l} = \int_{-1}^1 e^{-p_m(x+l)^2/b^2} \Phi_{ii'}(x) dx, \quad (10)$$

and δ , M , p_m , w_m , $m = 1, \dots, M$ are described in (2) and (3) for $\alpha = 1$.

We then use (7) instead of (6) to construct a multiresolution representation of the Poisson kernel (and bound state Helmholtz kernel) in [13, 14].

4. MADNESS

Multiwavelets were combined with low separation approximations using exponential functions for representation of functions and operators and implemented in dimensions 1, 2, 3, and 6 in our software package Multiresolution Adaptive Numerical Evaluation for Scientific Simulation (MADNESS). The initial prototype version used Python as the programming environment enabling the application code to be written at a very high level in terms of operators and functions, rather than the more common explicit manipulation of sparse lists of integrals and matrices. A wide range of electronic structure capabilities were developed using this framework. In addition, exploratory work has been conducted in fusion and fluid dynamics. The prototype runs in parallel on shared-memory computers using fork-and-join communicating between processes via files.

A new version is being implemented with a design goal of efficiently exploiting $O(10^5)$ processors for sufficiently large problems while retaining the high-level composition of applications. In effective one-particle (HF and DFT) calculations, there is one 3D molecular orbital per electron ($O(10^{2-3})$) each with an independent, adaptively-refined mesh. In effective two-particle (initially to be MP2) calculations there is one 6D function per electron pair, though at the MP2 level pairs are independent. Localization of the orbitals is used to reduce the overall scaling of HF and DFT to linear in the number of electrons. C++ is used for the high-level structure, using MPI to manage the coarse grain parallelism, and recursive traversal of trees via futures for finer grain parallelism within an SMP.

5. Current directions

The key to advancing the state of art in computing and, as a result, in quantum chemistry, materials science and physics, is computing in high dimensions. To this end, we are developing mathematical tools for making a transition from one-particle approximations (DFT, LDA, Hartree-Fock) to the two-particle approach. This requires computing in six spatial dimensions. Time dependent problems are also of interest and we are formulating several schemes with initial application to evolution of electronic systems in three and six dimensions. We are interested in generalizing results in [8] for computing (1) so that the family of functions $\{\phi_i^l(x_i)\}$ are Gaussians, $e^{-p_1 x_1^2} e^{-p_2 x_2^2} \dots e^{-p_n x_n^2}$, where the exponents p_1, p_2, \dots, p_n are allowed to be distinct in different directions. This will allow us to construct approximations to multiparticle Green's functions and spectral projectors. We also continue the work on general separated representations started in [6, 7].

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7. References

- [1] B. Alpert. A class of bases in L^2 for the sparse representation of integral operators. *SIAM J. Math. Anal.*, 24(1):246–262, 1993.
- [2] B. Alpert, G. Beylkin, D. Gines, and L. Vozovoi. Adaptive solution of partial differential equations in multiwavelet bases. *J. Comput. Phys.*, 182(1):149–190, 2002.
- [3] G. Beylkin, R. Coifman, and V. Rokhlin. Fast wavelet transforms and numerical algorithms. I. *Comm. Pure Appl. Math.*, 44(2):141–183, 1991. Yale Univ. Technical Report YALEU/DCS/RR-696, August 1989.
- [4] G. Beylkin, R. Coifman, and V. Rokhlin. Wavelets in numerical analysis. In *Wavelets and their applications*, pages 181–210. Jones and Bartlett, Boston, MA, 1992.
- [5] G. Beylkin, R. Cramer, G.I. Fann, and R.J. Harrison. Multiresolution separated representations of singular and weakly singular operators. *submitted*, 2004.
- [6] G. Beylkin and M. J. Mohlenkamp. Numerical operator calculus in higher dimensions. *Proc. Natl. Acad. Sci. USA*, 99(16):10246–10251, August 2002. <http://www.pnas.org/cgi/content/abstract/112329799v1>.
- [7] G. Beylkin and M. J. Mohlenkamp. Algorithms for numerical analysis in high dimensions. APPM preprint #519, Univ. of Colorado, February 2004. <http://amath.colorado.edu/pub/wavelets/papers/BEY-MOH2004P.pdf>, accepted for publication in *SIAM J. Sci. Comput.*
- [8] G. Beylkin and L. Monzón. On approximation of functions by exponential sums. *Appl. Comput. Harmon. Anal.*, 19(1):17–48, 2005.
- [9] S. F. Boys. The integral formulae for the variational solution of the molecular many-electron wave equations in terms of gaussian functions with direct electronic correlation. *Proceedings of the Royal Society of London Series a- Mathematical and Physical Sciences*, 258(1294):402, 1960. Proc. R. Soc. London Ser. A-Math.
- [10] D. Braess. Asymptotics for the approximation of wave functions by exponential sums. *J. Approx. Theory*, 83(1):93–103, 1995.
- [11] H. Cheng, L. Greengard, and V. Rokhlin. A fast adaptive multipole algorithm in three dimensions. *J. Comput. Phys.*, 155(2):468–498, 1999.
- [12] G.I. Fann, G. Beylkin, R.J. Harrison, and K. Jordan. Singular operators in multiwavelet bases. *IBM Journal of Research and Development*, 48(2):161–171, March 2004.
- [13] R.J. Harrison, G.I. Fann, T. Yanai, and G. Beylkin. Multiresolution quantum chemistry in multiwavelet bases. In P.M.A. Sloot et. al., editor, *Lecture Notes in Computer Science. Computational Science-ICCS 2003*, volume 2660, pages 103–110. Springer, 2003.
- [14] R.J. Harrison, G.I. Fann, T. Yanai, Z. Gan, and G. Beylkin. Multiresolution quantum chemistry: basic theory and initial applications. *J. Chem. Phys.*, 121(23):11587–11598, 2004. <http://amath.colorado.edu/pub/wavelets/papers/mrqc.pdf>.
- [15] W. Kutzelnigg. Theory of the expansion of wave functions in a Gaussian basis. *Internat. J. Quantum Chem.*, 51:447–463, 1994.
- [16] J.V.L. Longstaff and K. Singer. The use of gaussian (exponential quadratic) wave functions in molecular problems. ii. wave functions for the ground state of the hydrogen atom and of hydrogen molecule. *Proceedings of the Royal Society of London Series a- Mathematical and Physical Sciences*, 258(1294):421, 1960. Proc. R. Soc. London Ser. A-Math.
- [17] K. Singer. The use of gaussian (exponential quadratic) wave functions in molecular problems. i. general formulae for the evaluation of integrals. *Proceedings of the Royal Society of London Series a- Mathematical and Physical Sciences*, 258(1294):412, 1960. Proc. R. Soc. London Ser. A-Math.
- [18] T. Yanai, G.I. Fann, Z. Gan, R.J. Harrison, and G. Beylkin. Multiresolution quantum chemistry: Analytic derivatives for Hartree-Fock and density functional theory. *J. Chem. Phys.*, 121(7):2866–2876, 2004.
- [19] T. Yanai, G.I. Fann, Z. Gan, R.J. Harrison, and G. Beylkin. Multiresolution quantum chemistry: Hartree-Fock exchange. *J. Chem. Phys.*, 121(14):6680–6688, 2004.