DOE Computational Nanoscience Project: Integrated Multiscale Modeling of Molecular Computing Devices University of Colorado Final Report for the grant DE-FG02-03ER25583

Final report submitted by

Gregory Beylkin, Principal Investigator

Department of Applied Mathematics
526 UCB
University of Colorado at Boulder
Boulder, CO 80309-0526

Specific tasks of the original proposal

- Formulation of appropriate open and periodic boundary conditions within the multiwavelet approach.
- Comparison of various representations of functions and operators in the multiwavelet basis as a way of finding more effective ways of computing.
- Exploration of alternative types of multiresolution bases.
- Development of efficient representations for the Green's function at positive energies.
- Exploration of alternative implementations of the Green's function approach within the multiresolution framework.

Additional task of the non-cost extension

- Development of algorithms based on separated representations for *boundary value problems.*

Summary

Significant advances were made on all objectives of the research program. We have developed fast multiresolution methods for performing electronic structure calculations with emphasis on constructing efficient representations of functions and operators. We extended our approach to problems of scattering in solids, i.e. constructing fast algorithms for computing above the Fermi energy level. Part of the work was done in collaboration with Robert Harrison and George Fann at ORNL.

Specific results (in part supported by this grant) are listed here and are described in greater detail below.

1. We have implemented a fast algorithm to apply the Green's function for the free space (oscillatory) Helmholtz kernel. The algorithm maintains its speed and accuracy when the kernel is applied to functions with singularities.
2. We have developed a fast algorithm for applying periodic and quasi-periodic, oscillatory Green's functions and those with boundary conditions on simple domains. Importantly, the algorithm maintains its speed and accuracy when applied to functions with singularities.
3. We have developed a fast algorithm for obtaining and applying multiresolution representations of periodic and quasi-periodic Green's functions and Green's functions with boundary conditions on simple domains.
4. We have implemented modifications to improve the speed of adaptive multiresolution algorithms for applying operators which are represented via a Gaussian expansion.
5. We have constructed new nearly optimal quadratures for the sphere that are invariant under the icosahedral rotation group.
6. We obtained new results on approximation of functions by exponential sums and/or rational functions, one of the key methods that allows us to construct separated representations for Green's functions.
7. We developed a new fast and accurate reduction algorithm for obtaining optimal approximation of functions by exponential sums and/or their rational representations.

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5. Terry Haut, partially supported as a postdoc.

Results

We now detail the results stated in the summary.

1. We have constructed an approximation of the free space Green's function for the Helmholtz equation in such a way that the application of the resulting operator is split between the spatial and the Fourier domains, similar to Ewald method for evaluating lattice sums. Furthermore, we developed new quadratures appropriate for integration in the disk in the Fourier domain to account for the singularity of the Green's function. The contribution in the spatial domain requires a convolution with a small number of Gaussians with negative exponents and positive weights which fits well into our existing fast algorithm. The combination of approximation and new quadratures yields a fast algorithm for computing volumetric convolutions with this oscillatory Green's function in dimensions two and three. The algorithmic complexity effectively scales as $\mathcal{O}(k^d \log k + C(\log \epsilon^{-1})^d)$, where ϵ is selected accuracy, k is the number of wavelengths in the problem, d is the dimension, and C is a constant. Importantly, the algorithm maintains its efficiency when applied to functions with singularities. We note that our approximation differs from that used in the Fast Multipole Method and, as $k \to 0$, makes a smooth transition to the free space Green's function for the Poisson equation. The results have been published in [5].

2. We have developed a fast and accurate algorithm for computing convolutions with the quasi-periodic Helmholtz Green's function in two and three dimensions. This Green's function is defined by a conditionally convergent (lattice) sum. Although the main idea of our approach is similar to that of Ewald summation, in contrast to Ewald's method we use different means to interpret the sum. This provides an additional insight and a fast algorithm for its application. As in Ewald's method, we split the sum into two, one in the spatial domain and the other in the Fourier domain and both with exponential decay in their corresponding domains. In each domain we describe a method for fast convolution with combined algorithmic complexity $\mathcal{O}(k^d \log k)$, where k is the number of wavelengths in the problem and d is the dimension.
   Due to the exponential decay of the terms in both in the spatial and Fourier domains, we truncate them for a finite by arbitrary accuracy. As a result, our algorithm maintains its performance when applied to functions with singularities. We approximate the terms in the spatial domain as a sum of Gaussians having negative exponents and positive weights. Thus, to compute a convolution, we may use the multiresolution algorithm developed previously (and, in part, supported by this grant). In the Fourier domain, we use the Unequally Spaced Fast Fourier Transform (USFFT) to compute convolutions with the terms of Fourier sum. Our algorithm is applicable to volumetric problems and has computational complexity $\mathcal{O}(\kappa^d \log \kappa)$. We note that in the Fourier domain we use quadratures for bandlimited functions. The results have been published in [4].

3. We have developed a multiresolution representation of a class of integral operators satisfying boundary conditions on simple domains and constructed fast algorithms for their application. We have also elucidated some delicate theoretical issues related to the construction of periodic Green's functions for Poisson's equation.
   By applying the method of images to the non-standard form of the free space operator, we obtain lattice sums that converge absolutely on all scales, except possibly on the coarsest scale. On the coarsest scale the lattice sums may be only conditionally convergent and, thus, allow for some freedom in their definition. We use the limit of square partial sums as a definition of the limit and obtain a systematic, simple approach to the construction (in any dimension) of periodized operators with sparse non-standard forms. We illustrate the results on several examples in dimensions one and three: the Hilbert transform, the projector on divergence free functions, the non-oscillatory Helmholtz Green's function and the Poisson operator. Remarkably, the limit of square partial sums yields a periodic Poisson Green's function which is not a convolution. Using a short sum of decaying Gaussians to approximate periodic Green's functions, we arrive at fast algorithms for their application. We further show that the results obtained for operators with periodic boundary conditions extend to operators with Dirichlet, Neumann, or mixed boundary conditions. The results are available electronically [3].

4. We have implemented improvements to the algorithm based on separated multiresolution representations of Green's functions for the Poisson kernel and other non-oscillatory kernels and/or potentials. The speed up is about 3-5 times depending on the order of the multiwavelet basis (the more significant acceleration is for higher orders). This code is being used in the construction of solutions to the multiparticle Schrödinger equation, see [2, 6, 7].

5. We have constructed nearly optimal quadratures for the sphere that are invariant under the icosahedral rotation group. These quadratures integrate all $(N+1)^2$ linearly independent functions in a rotationally invariant subspace of maximal order and degree N. The nodes of these quadratures are nearly uniformly distributed and the number of nodes is only marginally more than the optimal $(N+1)^2/3$ nodes. Using these quadratures, we discretize the reproducing kernel on a rotationally invariant subspace to construct an analogue of Lagrange interpolation on the sphere. This representation uses function values at the quadrature nodes. In addition, the representation yields an expansion that uses a single function centered and mostly concentrated at nodes of the quadrature, thus providing a much better localization than spherical harmonic expansions. We show that this representation may be localized even further. We also describe two algorithms of complexity $\mathcal{O}(N^3)$ for using these grids and representations. Finally, we note that our approach is also applicable to other discrete rotation groups. The results are available in [1].

6. We have revisited the efficient approximation of functions by sums of exponentials or Gaussians in [8] and obtained new results and applications of these approximations. By using the Poisson summation to discretize integral representations of e.g., power functions $r^{-\beta}$, $\beta > 0$, we obtain approximations with uniform relative error on the whole real line. Our approach is applicable to a class of functions and, in particular, yields a separated representation for the function e^{-xy}. As a result, we obtain sharper error estimates and a simpler method to derive trapezoidal-type quadratures valid on finite intervals. We also introduce a new reduction algorithm for the case where our representation has an excessive number of terms with small exponents. As an application of these new estimates, we simplify and improve previous results on separated representations of operators with radial kernels. For any finite but arbitrary accuracy, we obtain new separated representations of solutions of Laplace's equation satisfying boundary conditions on the half-space or the sphere. These representations inherit a multiresolution structure from the Gaussian approximation leading to fast algorithms for the evaluation of the solutions. In the case of the sphere, our approach provides a foundation for a new multiresolution approach to evaluating and estimating models of gravitational potentials used for satellite orbit computations.
   We also considered the problem of reconstructing a compactly supported function with singularities either from values of its Fourier transform available only in a bounded interval or from a limited number of its Fourier coefficients. Our results are again based on several observations and algorithms in [8]. We avoid both the Gibbs phenomenon and the use of windows or filtering by constructing approximations to the available Fourier data via a short sum of decaying exponentials. Using these exponentials, we extrapolate the Fourier data to the whole real line and, on taking the inverse Fourier transform, obtain an efficient rational representation in the spatial domain. An important feature of this rational representation is that the positions of its poles indicate location of singularities of the function. We consider these representations in the absence of noise and discuss the impact of adding white noise to the Fourier data. We also compare our results with those obtained by other techniques. As an example of application, we consider our approach in the context of the kernel polynomial method for estimating density of states (eigenvalues) of Hermitian operators. We briefly consider the related problem of approximation by rational functions and provide numerical examples using our approach. The results have been published in [9, 10]

7. We have developed a fast and accurate reduction algorithm for computing optimal representations via exponentials and/or rational functions. When constructing rational approximations with an optimally small L^∞ error, we need to compute small con-eigenvalues and the associated con-eigenvectors of positive-definite Cauchy matrices. Specifically, given a rational function with n poles in the unit disk, a rational approximation with $m \ll n$ poles in the unit disk may be obtained from the mth con-eigenvector of an $n \times n$ Cauchy matrix, where the associated con-eigenvalue $\lambda_m > 0$ gives the approximation error in the L^∞ norm. Unfortunately, standard algorithms do not accurately compute small con-eigenvalues (and the associated con-eigenvectors) and, in particular, yield few or no correct digits for con-eigenvalues smaller than the machine round-off.
   We have develop a fast and accurate algorithm for computing con-eigenvalues and con-eigenvectors of positive-definite Cauchy matrices, yielding even the tiniest con-eigenvalues with high relative accuracy. The algorithm computes the mth con-eigenvalue in $\mathcal{O}\left(m^2 n\right)$ operations and, since the con-eigenvalues of positive-definite Cauchy matrices decay exponentially fast, we obtain (near) optimal rational approximations in $\mathcal{O}\left(n\left(\log \delta^{-1}\right)^2\right)$ operations, where δ is the approximation error in the L^∞ norm. We derive error bounds demonstrating high relative accuracy of the computed con-eigenvalues and the high accuracy of the unit con-eigenvectors. Finally, numerical tests on random (complex-valued) Cauchy matrices show that the algorithm computes all the con-eigenvalues and con-eigenvectors with nearly full precision. This research started during the no-cost extension of this grant and results may be found in [11].

Publications

1. G. Beylkin, C. Kurcz and L. Monzón, "Fast Convolution with the free space Helmholtz Green's function", Journal of Computational Physics , 228, (8) (2009) 2770–2791

2. G. Beylkin, M. J. Mohlenkamp and F. Perez, Preliminary results on approximating a wavefunction as an unconstrained sum of Slater determinants, Proc. Appl. Math. Mech., 7, (2007)

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8. Beylkin and L. Monzón, "Nonlinear inversion of a bandlimited Fourier transform", Applied and Computational Harmonic Analysis, 27, (2009) 351–366

9. C. Ahrens and G. Beylkin, "Rotationally invariant quadratures for the sphere", Proceedings of the Royal Society A, 465, (2009) 3103–3125

10. G. Beylkin and L. Monzón, "Approximation by exponential sums revisited", Applied and Computational Harmonic Analysis, 28, (2010) 131–149

11. T. S. Haut and G. Beylkin, "Fast and accurate con-eigenvalue algorithm for optimal rational approximations", arXiv:1012.3196v2 [math.NA], 2011.

12. G. Beylkin, G. Fann, R. J. Harrison, C. Kurcz, and L. Monzón. "Multiresolution representation of operators with boundary conditions on simple domains", Applied and Computational Harmonic Analysis, 2011. Available at http://dx.doi.org/10.1016/j.bbr.2011.03.031.

Presentations

1. Beylkin, G., "Fast algorithms for adaptive application of integral operators in high dimension", The 2007 John H. Barrett Memorial Lectures, April 28, 2007

2. Beylkin, G., "Nonlinear inversion of bandlimited Fourier transform and discrete transforms for bandlimited functions in a disk ", IPAM, Lake Arrowhead, June 10-15, 2007

3. Beylkin, G., Martin J. Mohlenkamp and Fernando Pérez, "Toward Solving the Multiparticle Schrodinger Equation via an Unconstrained Sum of Slater Determinants", NWChem, Jan 25, 2007

4. Beylkin, G., "Fast algorithms for adaptive application of integral operators in high dimension", University of North Carolina at Chapel Hill, February 2, 2007

5. Beylkin, G.; Mohlenkamp, M. J.; Pérez, F., "Preliminary results on approximating a wavefunction as an unconstrained sum of Slater determinants", ICIAM Meeting, Minisymposium "High-dimensional analysis meets scientific computing", Zurich, Switzerland, July 2007

6. Beylkin, G., "Separated Representations and Nonlinear Approximations for Fast Algorithms in High Dimensions", ICIAM Meeting, Minisymposium "Numerical multilinear algebra: a new beginning", Zurich, Switzerland, July 2007

7. F. Pérez, "Adaptive Application of Green's Functions: Fast algorithms for integral transforms, with a bit of QM", U. of Michigan at Ann Arbor, 2007.

8. F. Pérez, “Modern algorithms in mathematical research, parallelism and languages: The intersection of theoretical and practical issues”, Invited talk at Cyber-Enabled Discovery and Innovation workshop, NSF headquarters, October 2007.

9. F. Pérez, “Adaptive Application of Green’s Functions: Fast algorithms for integral transforms, with a bit of QM”, University of Michigan at Ann Arbor, 2007.

10. Beylkin, G., “Fast algorithms for adaptive application of integral operators in high dimension”, Snapshots of Applied Mathematics SIAM, Great Lakes Section, April 12, 2008

11. Monzón, L., “Approximation by exponential sums revisited”, SIAM Annual meeting, San Diego, California, July 10, 2008

12. Beylkin, G., “On approximation of operators by Gaussians”, IPAM, UCLA, February 12, 2009

13. Beylkin, G., “Fast algorithms for applying multidimensional Green’s functions”, SIAM Conference on Computational Science and Engineering (CSE09), Miami, FL, March 2, 2009.

14. Beylkin, G., “Nonlinear approximations and multiresolution algorithms in high dimensions”, Twenty years of wavelets, DePaul University, Chicago, May 16, 2009

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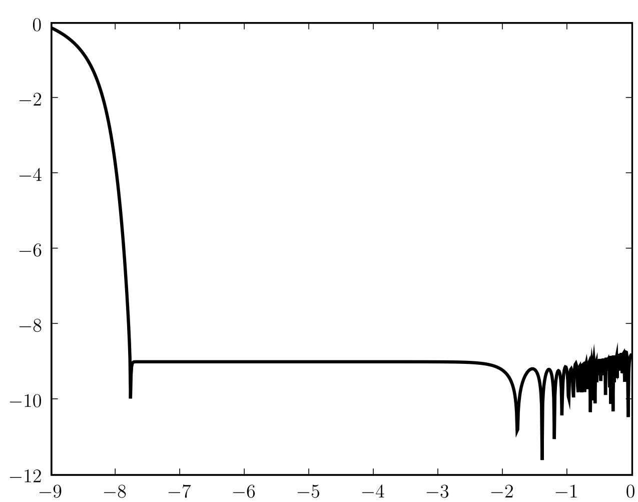

(a)

Figure 1: Absolute error (as a function of radius using $\log_{10} - \log_{10}$ scale) of approximating the Green’s function in dimension $d = 2$, $\frac{i}{4}H_0^1(kr)$, with $k = 50\pi$ and $\epsilon \approx 10^{-8}$(see [5] for details).

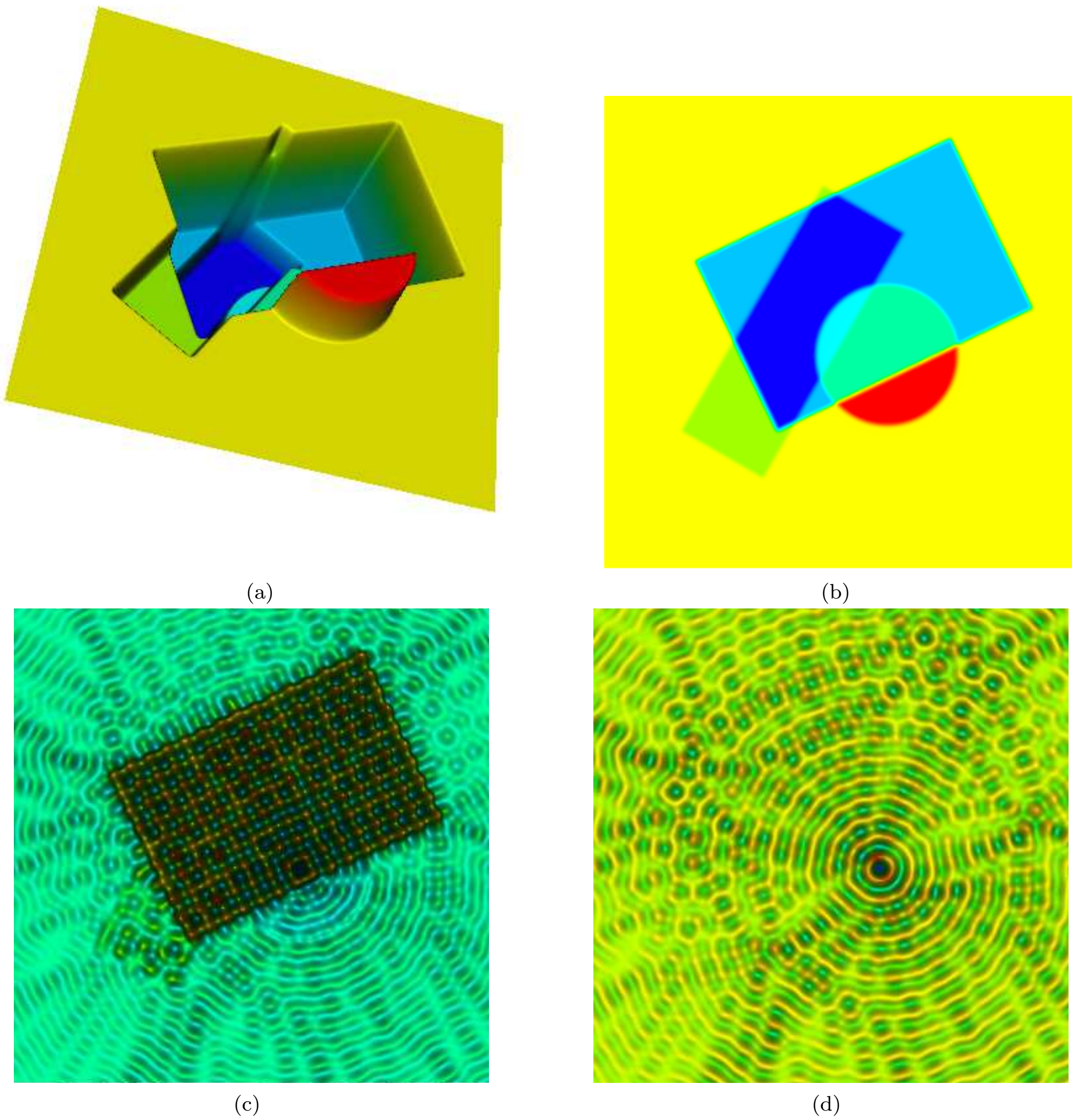

Figure 2: Convolution with the Green's function $\frac{i}{4}H_0^1(kr)$ with $k = 50\pi$, where the different views of the discontinuous function are shown in Figures (a) and (b). We display the real part (c) and imaginary part (d) of the result (see [5] for details).

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