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RPPR Final Report
as of 24-Apr-2019

Agency Code:

Proposal Number: 69796CHII **Agreement Number: W911NF-16-1-0570**
INVESTIGATOR(S):

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Report Date: 31-Oct-2017 Date Received: 28-Jan-2019
Final Report for Period Beginning 01-Nov-2016 and Ending 31-Jul-2017
Title: Canonical Tensors Applied to Ab Initio Electronic Structure: Linear Scaling for Metallic Systems
Begin Performance Period: 01-Nov-2016 **End Performance Period:** 31-Jul-2017
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Submitted By: Kathleen Calhoun Email: calhounka@tsu.edu
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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 199 **STEM Participants:** 199

Major Goals: An ab initio electronic structure algorithm capable of achieving linear scaling for metallic systems, and utilizing hundreds of thousands of Cardinal Sine basis elements on dual core processor with 16Gb memory, is presented. This is achieved by the implementation of a dimensionally separated Canonical Tensor Representation (CTR) of the Hamiltonian Matrix, and associated Density Matrix. Rank reduction of the Canonical Tensor Representation, required for matrix multiplies, was first developed by G. Beylkin et. al. (SIAM Journal on Scientific Computing 26 (2005) 2133). This algorithm allows us to represent the full six-dimensional tensor (matrix) as three separated rank two-dimensional tensors, where we have extended Beylkins method via the use of the Frobenius norm for the two-dimensional tensors. A significant advantage is achieved with the Cardinal Sine basis via the use of the Gaussian-Sinc integral technology, developed by J. Jerke et. al. (J Chem Phys 143 (2015) 064108), which allows us to construct the Hamiltonian in a natural canonical tensor-product of rank-2 tensors. By separating space into three one- dimensional forms (N3 --> N), linear scaling is attained with dense matrix representations. The matrix-matrix multiply operations are now linear because the scaling of the matrix- matrix multiply is a cube of a cubic root of the number of basis elements and the rank of the canonical tensors are insensitive to the system size. Solving for a density matrix, or canonical density, is solved with aid of the Spectral Projection algorithm (SP2) technologies developed by Niklasson et. al. (Physical Review B 66 (2002) 155115). Efficient storage of density matrices into dimensionally separated ‘primitives’ and extremely fast builds of the Fock matrices allow for novel Self Consistent Field (SCF) acceleration. The energy represented in the primitive basis is minimized linearly with exponential cost functions associated with idompotency. A dozen SCFs converge metallic systems with up to 357,911 basis elements for lithium. To conclude, the proposed method of dimensional space separation with canonical tensors achieves efficiency for even metallic systems, with excellent accuracy, and in a reasonable computational effort.

Accomplishments: 1. Digital Basis span broad bonding regions
2. Gaussians span to wave function cusps
B. Derivation of Separated dimensional (CTR) Quantum Operators
3. Natural way to separate dimensions with analytic formula
4. N3 -> N, with N scaling for relevant computations
1. Cubic Speedup compared to normal algorithms
5. Implementation of Alternating Least Squares to maintain sparse rep of vectors under matrix-vector multiply
6. Extension of Discrete Variable Representation to full basis representations(FBR)
1. Applicable to metallic systems b/c of FBR quantum operators
7. Electron-Electron interaction (2-body matrix)

8. Screened Electron-Electron interaction (2-body matrix)
9. Electron-Nuclear interaction (1-body matrix)
C. Exact band-limited computation of a dozen low lying 2-body wave functions of
10. helium
11. molecular hydrogen (geometries including ground state through dissociation)
12. Final computation converged 101 lattice grid elements on a side: tera basis
13. Exact correlation written into 2-body wave functions
[more recently and proceeding after grant]
D. Exact band-limited computation of the Thomas-Fermi model with 2, 3, and 4 electrons per periodic cell
14. Different perspective on Thomas Fermi and relevant to Uniform Electron Gas
15. Direct and exact computation of quantum metallic behaviors

Training Opportunities: Attend the 2017 Chemistry Division and ARO Program Review in Research Triangle Park, NC- August 6-11, 2017

From: "Parker, James K CIV USARMY RDECOM ARL (US)"
<james.k.parker30.civ@mail.mil>
To: "Jerke, Jonathan" <jonathan.jerke@ttu.edu>
Subject: FW: New ARO program review requirement: 7-11 August
(UNCLASSIFIED)
CLASSIFICATION: UNCLASSIFIED
Hi Jonathan,
You are invited to attend the 2017 Chemistry Division program review. The
meeting is intended to bring together PIs from all of the ARO chemistry programs to discuss their research. All PIs will give a 30-minute presentation of their
research. The dates for the meeting are 7 -11 August 2017. The location will be Research Triangle Park, NC. Financial support to attend this meeting can come directly from your ARO grant, if you so desire. Please see the below message and the attached documents for further information.
best regards,
Jim

Results Dissemination: The original intent of the Army Research Office grant was to compute excited state correlated systems with time dependence. All basic principles behind this kind of goal have been achieved. The foundation of the work is the basis, developed in the preceding few years (2015). The work published in early 2018, “Two-Body Schrödinger Wave Functions in a Plane-Wave Basis Via Separation of Dimensions” by Jerke and Poirier demonstrated direct and exact solves of Schrödinger’s equation for more than 1-body in a plane-wave basis. Attaining few-electron wave functions of dozens of excited states and the electronic ground state, which is a common occurrence now on our systems, could easily be made time dependent by any number of trivial approaches.

Honors and Awards: Podium presentation at the ARO Program Review in Research Triangle Park, NC August 6-11, 2017.

Protocol Activity Status:

Technology Transfer: See attached journal.

PARTICIPANTS:

Participant Type: PD/PI
Participant: Christopher Tymczak
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Other (specify)
Participant: Bobby L Wilson
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Jonathan Jerke
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
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National Academy Member: N
Other Collaborators:

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
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Volume: 148 Issue: 10 First Page #: 104101
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Publication Location:
Article Title: Two-body Schrodinger wave functions in a plane-wave bassis via separation of dimensions
Authors: Jonathan Jerke, and Bill Poirier
Keywords: Plane-wave
Abstract: Using a combination of ideas, the ground and several excited electronic states of the helium atom and the hydrogen molecule are computed to chemical accuracy—i.e., to within 1–2 mhartree or better. The basic strategy is very different from the standard electronic structure approach in that the full twoelectron six-dimensional (6D) problem is tackled directly, rather than starting from a single-electron Hartree-Fock approximation. Electron correlation is thus treated exactly, even though computational requirements remain modest. The method also allows for exact wave functions to be computed, as well as energy levels. From the full-dimensional 6D wave functions computed here, radial distribution functions and radial correlation functions are extracted—as well as a 2D probability density function exhibiting antisymmetry for a single Cartesian component.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

Final Grant Report

Army Research Office grant W911NF-16-1-0570

Title

Canonical Tensors Applied to Ab Initio Electronic Structure: Linear Scaling for Metallic Systems

Abstract

An ab initio electronic structure algorithm capable of achieving linear scaling for metallic systems, and utilizing hundreds of thousands of Cardinal Sine basis elements on dual core processor with 16Gb memory, is presented. This is achieved by the implementation of a dimensionally separated Canonical Tensor Representation (CTR) of the Hamiltonian Matrix, and associated Density Matrix. Rank reduction of the Canonical Tensor Representation, required for matrix multiplies, was first developed by G. Beylkin et. al. (SIAM Journal on Scientific Computing 26 (2005) 2133). This algorithm allows us to represent the full six-dimensional tensor (matrix) as three separated rank two-dimensional tensors, where we have extended Beylkins method via the use of the Frobenius norm for the two-dimensional tensors. A significant advantage is achieved with the Cardinal Sine basis via the use of the Gaussian-Sinc integral technology, developed by J. Jerke et. al. (J Chem Phys 143 (2015) 064108), which allows us to construct the Hamiltonian in a natural canonical tensor-product of rank-2 tensors. By separating space into three one- dimensional forms (N3→N), linear scaling is attained with dense matrix representations. The matrix-matrix multiply operations are now linear because the scaling of the matrix- matrix multiply is a cube of a cubic root of the number of basis elements and the rank of the canonical tensors are insensitive to the system size. Solving for a density matrix, or canonical density, is solved with aid of the Spectral Projection algorithm (SP2) technologies developed by Niklasson et. al. (Physical Review B 66 (2002) 155115). Efficient storage of density matrices into dimensionally separated ‘primitives’ and extremely fast builds of the Fock matrices allow for novel Self Consistent Field (SCF) acceleration. The energy represented in the primitive basis is minimized linearly with exponential cost functions associated with idompotency. A dozen SCFs converge metallic systems with up to 357,911 basis elements for lithium. To conclude, the proposed method of dimensional space separation with canonical tensors achieves efficiency for even metallic systems, with excellent accuracy, and in a reasonable computational effort.

Granted to

Texas Southern University

CJ Tymczak and executed by Bobby Wilson
Completed by Jonathan Jerke

Completed Key Points

A. Gaussian-Sinc Basis
   1. Digital Basis span broad bonding regions
   2. Gaussians span to wave function cusps

B. Derivation of Separated dimensional (CTR) Quantum Operators
   3. Natural way to separate dimensions with analytic formula
   4. N^3 -> N, with N scaling for relevant computations
      1. Cubic Speedup compared to normal algorithms
   5. Implementation of Alternating Least Squares to maintain sparse rep of vectors under matrix-vector multiply
   6. Extension of Discrete Variable Representation to full basis representations(FBR)
      1. Applicable to metallic systems b/c of FBR quantum operators
   7. Electron-Electron interaction (2-body matrix)
   8. Screened Electron-Electron interaction (2-body matrix)
   9. Electron-Nuclear interaction (1-body matrix)

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   10. helium
   11. molecular hydrogen (geometries including ground state through dissociation)
   12. Final computation converged 101 lattice grid elements on a side: tera basis
   13. Exact correlation written into 2-body wave functions

[more recently and proceeding after grant**]**

D. Exact band-limited computation of the Thomas-Fermi model with 2, 3, and 4 electrons per periodic cell
   14. Different perspective on Thomas Fermi and relevant to Uniform Electron Gas
   15. Direct and exact computation of quantum metallic behaviors

Papers

J. Jerke, Y. Lee, and C. J. Tymczak, J Chem Phys 143 (2015) 064108.
J. Jerke and P. Bill, J Chem Phys 148 (2018) 104101.
[more recently and proceeding after grant**]**
J. Jerke, J. Karwowski, and B. Poirier, Molecular Phys (2018) 1.
J. Jerke, E. Bittner, and B. Poirier, (in press) (2019).

Software

A. PERSEUS house software written in C with OpenMPI
B. ANDROMEDA available on Github.com and written in C with OpenMP

Report

CHEMICAL SCIENCES DIVISION , James Parker

The original intent of the Army Research Office grant was to compute excited state correlated systems with time dependence. All basic principles behind this kind of goal have been achieved. The foundation of the work is the basis, developed in the preceding few years (2015). The work published in early 2018, "Two-Body Schrödinger Wave Functions in a Plane-Wave Basis Via Separation of Dimensions" by Jerke and Poirier demonstrated direct and exact solves of Schrödinger's equation for more than 1-body in a plane-wave basis. Attaining few-electron wave functions of dozens of excited states and the electronic ground state, which is a common occurrence now on our systems, could easily be made time dependent by any number of trivial approaches.

There are several approaches into the many-electron limit:

A. DFT augmented with a few-body parameter
B. Mazziotti approach to many-electrons via Linear Programming in few-body basis
C. Few-Body Hartree Fock + MP2
D. RPA like approach with geminals (instead of just orbitals)

Our current work resides around understanding the DFT augmentation with a few-body parameter as discussed by Gill and Loos (2012). The Mazziotti approach has been investigated and met with him in 2018 to introduce my work to him (Mazziotti is also supported by the ARO). The Few-Body Hartree Fock theory has been written down by Gonis and Schulthess (1998) and would be a significant improvement over the 1-body Hartree Fock proposed previously. There is also a rich history of works associated with geminals—2-body orbitals—and Random Phase Approximation (RPA) dating back to the 1970's that was previously impossible without direct representation of 2-body wave functions.

After great efforts and the loss of Tymczak, Jerke abandoned the effort to solve 1-body Hartree Fock for metallic systems. Clearly a fast algorithm is desirable but the difficulties of its implementation and lack of exact correlation lead to a refocusing on the core CTR technologies. Essentially retreating into to higher dimensions to solve novel problems with direct and exact linear solves.

We now understand that few-body bases are fundamentally superior to 1-body bases and can reconsider the many-electron limit with any of the listed technologies. We know how to map 'down' to the 1-body full CI solution: the current gold standard. Our experience is that 2-body wave functions can be achieved in linear scaling (order volume) computation complexity and convergence of extreme bases is nearly assured using the band-pass staging algorithm developed during this time.

We have continued efforts to consider metallic systems via solving the exact Thomas-Fermi model for few-electrons. As well as exact and direct solves of atomic/periodic Lithium are in the works. The key point of metallic systems is the FBR of the matrices, which is only required for metals. The CTR FBR quantum operator technology is completed and published.

The majority of the work during this grant covered the coding requirements to complete **ANDROMEDA**: *a few electron plane-wave calculator*. A lot of expertise was found with my collaboration, Bill Poirier at Texas Tech University, as well as some of the inspiration for the helium work. Andromeda has been since released under a GPL3.0 license on GitHub.com. Current capabilities include mixed spatial and periodic computations of up to 4 electrons with wave function outputs. There has also been the development of a free-iOS app called Quantum Galaxies AR available on the APPLE Store. This work has been completed to increase the interest in this analytic data science field where a tera-bit of data can be processed in an iOS device every second using CTR technologies.

After the grant ended, Jerke continued work under a Welch Grant at Texas Tech University. Proceeding directly from this ARO grant was validation of general 2-body pair-wise quantum operators (2018) and solution of the few-body Thomas-Fermi model (2019). This last paper acknowledges continued support from the Army Research Office.