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# RPPR Final Report
as of 07-Jan-2019

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**Distribution Statement:** 1-Approved for public release; distribution is unlimited.

**STEM Degrees:** 0 **STEM Participants:** 0

**Major Goals:** The overarching objective of the proposed work is to develop the formulation and implementation of large-scale time dependent DFT (TDDFT) calculations. The aim is to develop methods for both pseudopotential as well as all-electron calculations. The specific goals include:
1. Development of a real-space formulation of TDDFT that is applicable to both psuedopotential as well as all-electron calculations.
2. Finite element discretization of the formulation with basis adaptation informed by numerical analysis.
3. An adaptive coarse-graining of the temporal evolution of the wave functions informed by numerical analysis.
4. Development of subspace projection techniques that will significantly improve the efficiency and computational complexity of TDDFT calculations.
5. Develop a scalable implementation of the algorithms on massive parallel computing platforms.

**Accomplishments:** The following have been accomplished over the course of the project.

1. As the overarching goal is to develop the TDDFT calculations for both pseudopotential as well as all-electron calculations, we developed a framework to efficiently conduct all-electron calculations. To this end, we developed an enriched finite-element basis, which enriches the classical finite-element space with enrichment functions that are computed for single atom Kohn-Sham orbitals. We conducted studies to ascertain the convergence, accuracy and efficiency afforded by the enriched finite element basis. Using ground-state calculations as a metric, we demonstrated that the enriched FE basis significantly outperforms existing widely used gaussian-type basis functions (with computational efficiency that is five- to ten-fold greater than using state of the art all-electron codes). Ground-state calculations on systems approaching 10,000 electrons have been demonstrated. This work was published in: Kanungo and Gavini, Phys. Rev. B 95 035111 (2017).

2. We developed a computationally efficient approach to solve the time-dependent Kohn-Sham equations in real-time using higher-order finite-element spatial discretization, applicable to both pseudopotential and all- electron calculations. To this end, we develop an a priori mesh adaption technique, based on the semi-discrete (discrete in space but continuous in time) error estimate on the time-dependent Kohn-Sham orbitals, to construct a close to optimal finite-element discretization. Subsequently, we obtain the full-discrete error estimate to guide our choice of the time-step. We employ spectral finite-elements along with Gauss-Legendre- Lobatto quadrature to render the overlap matrix diagonal, thereby simplifying the inversion of the overlap matrix that features in the evaluation of the discrete time-evolution operator. We use the second-order Magnus operator as the time-evolution operator in all our calculations. Furthermore, the action of the discrete Magnus operator, expressed as exponential of a matrix, on

the Kohn-Sham orbitals is obtained efficiently through an adaptive Lanczos iteration. We observe close to optimal rates of convergence of the dipole moment with respect to spatial and temporal discretization, for both pseudopotential and all-electron calculations. We demonstrate a staggering 100-fold reduction in the computational time afforded by higher- order finite-elements over linear finite-elements, for both pseudopotential and all-electron calculations. We present comparative studies, in terms of accuracy and efficiency, of our approach against finite-difference based discretization for pseudopotential calculations, and demonstrate significant computational savings when compared to the finite-difference method (OCTOPUS). We also demonstrate the competence of higher-order finite-elements for all-electron benchmark systems. Lastly, we observe good parallel scalability of the proposed method on many hundreds of processors. This work has been submitted to the Journal of Computational Physics, and is under review. The preprint is available on arXiv, arXiv:1810.13130 .

3. We have developed a subspace projection method for an efficient solution of the TDDFT problem by using the eigenspaces of the instantaneous Hamiltonians (computed from efficient Chebyshev acceleration techniques) from previous time steps. The choice of the subspace and the frequency with which the subspace is updated is being guided by numerical analysis. Using this subspace projection technique, on the benchmark problems we studied, we achieve a staggering 10-fold computational speed up as compared to conventional temporal evolution. This is due to: (i) the significantly smaller subspace size required in the present approach in comparison to the Krylov subspace used in conventional temporal evolution; (ii) the subspace only needs to be updated infrequently in comparison to the evolution of the Kohn-Sham wavefunctions. These results are very promising and provides a path towards conducting large-scale TDDFT calculations. A manuscript based on these ideas and results is under preparation and will be submitted shortly.

4. The numerical implementation of the real-space discretization of the TDDFT problem and the algorithms developed in this work have been implemented in a parallel computing framework. Good parallel scalability has been demonstrated on up to 1000 processors. In the near future, we will merge the aforementioned developments in TDDFT to the massively parallel open source DFT-FE code, which will enable a-posteriori spatial adaption, and further improve parallel scalability and efficiency.

**Training Opportunities:** A graduate student has received multi-disciplinary training in electronic structure calculations, numerical methods, numerical analysis and scientific computing.

**Results Dissemination:** Kanungo, B., Gavini, V., Large-scale all-electron density functional theory calculations using an enriched finite element basis, Phys. Rev. B 95, 035112 (2017).

Kanungo, B., Gavini, V., Real-time time-dependent density functional theory using higher order finite element methods, submitted to J. Comput. Phys. (2018). arXiv preprint arXiv:1810.13130

Kanungo, B., Gavini, V., A subspace projection method for large-scale real-time TDDFT calculations, under preparation.

**Honors and Awards:** Bikash Kanungo received the Michigan Institute for Computational Discovery and Engineering Scientific Computing fellowship for his work on numerical methods for TDDFT. Besides the recognition of his scholarship, the fellowship amount of $4,000 can be used towards furthering his research and travel to conferences.

**Protocol Activity Status:**

**Technology Transfer:** We have regular conference calls with scientists at the U.S. Army Research Laboratory in Aberdeen to discuss the developments in this project. Further, PI visited U.S. Army Research Laboratory in Aberdeen and Adelphi (in October 2018) to present the developments from his group in electronic structure calculations, which included developments from this project. There is interest from some groups in Adelphi to use these developments in TDDFT to understand photo-enhanced reaction mechanisms. We are in active discussions to collaborate on these studies.

**PARTICIPANTS:**

**Participant Type:** PD/PI
**Participant:** Vikram Gavini

**Person Months Worked:** 7.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

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Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

**Participant Type:** Graduate Student (research assistant)
**Participant:** Paavai Pari
**Person Months Worked:** 2.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Nothing to report in the uploaded pdf (see accomplishments).