DE-SC0016415: Final Report

Support of A Summer School Workshop and Workshop Focused on Theory and Applications of Time-Dependent Density Functional Theory

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Abstract

The first US-based summer school and workshop on Time-Dependent Density Functional Theory (TDDFT) was held July 11-21, 2017 in Telluride, CO. This grant provided funding to enable 33 students to attend the school, specifically with lodging and registration fee reductions. TDDFT is increasingly used in computational molecular and materials science to calculate electronic-excitation spectra and dynamics in a wide variety of applications, including photocatalysis, photo-controlled bond dissociation, and light-induced charge transfer. Software development in this community targets multiple software packages, many of which are open source, such as octopus, NWchem and Qb@ll, which are the ones our school focused on. The goal of this first iteration was to create a home for a national community of scholars, including users and developers, with a deep understanding of TDDFT, its capabilities, limitations, and high-performance computing context. We used this opportunity to explore interest in such an event in the future and based on overwhelmingly positive feedback from students and teachers, we intend to hold a similar school+workshop every two years in the US, in order to maintain the high level of interest that we witnessed and the enthusiasm amongst participants.

Summary of School and Workshop Activities

The school consisted of theory lectures in the morning followed by hands-on practical sessions in the afternoon.

The morning lectures were 50 min each, with 20 minutes for questions and discussion. This was a successful format because it turned out to be highly interactive, with many questions by the students. Topics covered were: Fundamentals of DFT, Fundamentals of TDDFT, Linear Response, Excitations in Solution, Excitations in the Solid-State, Charge-Transfer and Optimally-Tuned Hybrids, Real-Time TDDFT, time-dependent current DFT, and Non-Adiabatic Electron-Ion Dynamics. All lecturers uploaded their materials to a web site that all students were able to access. Each instructor (Kieron Burke, Mark Casida, Christine Isborn, Neepa Maitra, Shane Parker, Lucia Reining, Andre Schleife) gave two lectures.

The first two afternoon hands-on sessions, led by Niri Govind and Ken Lopata, focused on the NWChem code, but also used python, octave, and gnuplot. Students learned how to set up an input file to calculate excitation energies using linear response TDDFT and also perform electron dynamics using real-time TDDFT. All students were able to have a working code for modeling charge-transfer excitations with different functionals and peak-transfer from resonant laser-induced excitation. The second two afternoon sessions, led by Alberto Castro and Xavier Andrade, focussed on the Qb@ll and Octopus codes, in order to complement the local-orbital aspect of the first two sessions with the plane-wave approach to describe Kohn-Sham states. Students applied a laser field to a molecule and analyzed the resulting electron dynamics, performed Ehrenfest dynamics to model photodissociation, and simulated a high harmonic generation spectrum. All hands-on sessions were run on NERSC supercomputers, in order to provide students with a “real-world” experience of how to run simulations in a high-performance-computing context. To this end, we successfully applied for and used NERSC training accounts.

On the final day all instructors were present for a one-hour panel discussion with the students. This discussion was remarkably lively; a large fraction of the students remained engaged with extremely pertinent questions throughout the discussion, on prospects of advances in theoretical functional development as well as practical application of the theory. The discussion even went over the scheduled hour, even after the full week of lectures and time for questions.

We held a poster session which enabled students to directly discuss their on-going work with the lecturers and with other students. The breakfasts, lunches, and dinners provided further opportunity for discussion. In particular, the breakfasts, being provided by TSRC on-site, allowed many participants to interact and get to know each other. For the workshop, lunches were provided on-site, enabling extensive small-group discussions of the research presented in the workshop talks and effectively connecting the students who stayed for the workshop with the workshop speakers.

The workshop topics spanned recent developments and applications in TDDFT, from molecules to solids, and from linear response to strong fields. The overall salient observation was the extensive range of applications of TDDFT to quite complex situations, and the depth of the analysis, both in terms of the computational efficiency as well as functional accuracy. For example, with electron scattering, we heard talks on dynamical features in the exact time-dependent exchange-correlation potential in electron-molecule scattering, and, at the other extreme, results on scattering in warm dense plasmas. The advances in stopping power calculations over the recent years was notable for the size and scope of what is possible today; when is it important to include the vibrations of the lattice and when can they be ignored, when must we go beyond the pseudopotential approximation, and what is the effect of defects in semiconductors on stopping power. Then again on the more fundamental side, there was a talk on whether a long-time thermal equilibrium for TDDFT can be defined. Effects arising from quantized radiation fields coupled to molecules were discussed, with new density-functional methods that bridge quantum optics with quantum chemistry, and recent advances in multiscale methods for strong classical fields coupled to electron-ion dynamics in materials were presented. New developments of TDDFT in solvents were presented, in particular careful

examination of the effects of coupling explicit quantum solvent with a polarizable continuum. The full range of talks and topics is evident in the talk titles presented below. One afternoon session was devoted to talks in honor of Giovanni Vignale's 60th birthday. Applications of TDDFT to such complex situations, together with analysis of the fundamental aspects, beyond a simple calculation of spectra of intermediate-size molecules or periodic solids, exemplifies the progress in the field in recent years.

Participants of School and Workshop

The TDDFT school drew about 60 applicants from around the world, out of whom we selected 33 based on their promise for a career in science and their research field of interest. We strove for a balance of students from large and small institutions, from groups with established and early-career research advisors, and from a range of experience levels. Of the 33 students, 8 were women.

Student Participants, Summer School:

1. Cheng-Wei Lee, University of Illinois at Urbana-Champaign
2. Niraj Nepal, Temple University
3. Christopher Myers, University at Albany
4. Ravithree Senanayake, Kansas State University
5. John Philbin, UC Berkeley
6. Guo Chen, UC Irvine
7. Daniel Burrill, University of Pittsburgh
8. Edwin Quashie, Lawrence Livermore National Laboratory
9. Nathan Koocher, Northwestern University
10. Zuxin Jin, University of Pennsylvania
11. Ying Zhu, Ohio State University
12. Adam Bruner, Lousiana State University
13. Francisca Sagredo, University of California Irvine
14. Hemanadhan Myneni, University of Delaware
15. Sudheer Peddathimmareddy, Rutgers University
16. Dominik Maximilian Juraschek, ETH Zurich, Switzerland
17. Alexander Kohn, MIT
18. Imon Mandal, Tata Institute of Fundamental Research (TIFR), Mumbai
19. Dillon Yost, Univ. North Carolina, Chapel Hill
20. Rodrigo Freitas, UC Berkeley
21. Irene Metz, University of Iowa
22. Jon Bender, UT Austin
23. Andrew Sifain, Los Alamos National Laboratory
24. Amelia Fitzsimmons, Oak Ridge National Laboratory
25. Arthur De Vos, Ghent University

26. Feng Wu, UC Santa Cruz
27. Jane Herriman, Caltech
28. Yanal Oueis, Purdue University
29. Graeme Gossel, Hunter College
30. Jacob Wilmer, UC Merced
31. Edward Pluhar, University of Missouri
32. Aliya Mukazhanova, Boston University
33. Matthew Anderson, University of Missouri

The Excited States workshop had 34 speakers. Some of the speakers were students from the summer school that remained for the workshop. The talks spanned many research areas, and the titles are given below.

Excited States Workshop Participants, Affiliations, and Talk Titles:

1. Kieron Burke, Dept of Chemistry, UC Irvine: *Excitation energies from ensemble DFT*
2. Yasumitsu Suzuki, Dept of Physics, Tokyo University of Science: *Exact time-dependent exchange-correlation potentials in electron scattering processes*
3. Prineha Narang, Materials Science, Harvard University: *Excited state and Quantum-engineered Materials*
4. Graeme Gossel, Dept. of Physics and Astronomy, Hunter College CUNY : *Exact factorization of the electron-nuclear wavefunction: properties, approximations, and applications*
5. Giovanni Vignale, Dept of Physics and Astronomy, University of Missouri: *Berry curvature and excited states at the boundary of an electron liquid*
6. Carsten Ullrich, Dept of Physics and Astronomy, University of Missouri: *Spin waves in chiral two-dimensional electron systems*
7. Florian Eich, Max-Planck Institute Hamburg: *Charge and Energy Transport at the Nanoscale: A Density Functional Perspective*
8. Irene D'Amico, Dept of Physics, York University, UK: *Using DFT concepts and tools for quantum thermodynamics*
9. Christine Isborn, Dept of Chemistry, U.C. Merced: *Excited States in Solution: Towards More Accurate Absorption Spectra*
10. Matthieu Verstraete, Dept of Physics, University of Liege, Belgium: *Long-range dispersion forces between molecules subject to attosecond pulses from ab initio calculations*
11. Imon Mandal, Tata Institute of Fundamental Research (TIFR), Mumbai: *Photoinduced Charge Transfer transitions Associated with Charged Amino Acids:TDDFT Reveals a new Spectral Marker for Proteins*
12. Andrew Baczewski, Center for Computing Research, Sandia National Laboratory: *Warm Dense Matter: Opportunities and challenges for TDDFT*
13. André Schleife, Dept of Materials Science and Engineering, University of Illinois at Urbana-Champaign: *Excited-electron dynamics near surfaces: Particle radiation in real-time TDDFT*

14. Xavier Andrade-Valencia, Lawrence Livermore National Laboratory: *Non-linear effects in conductivity*
15. John Parkhill, Dept. of Chemistry, University of Notre Dame: *Pumping and Relaxation in TDDFT: Patches and problems*
16. David Strubbe, Dept. of Physics, U.C. Merced: *Excited-state forces in TDDFT and the Bethe-Salpeter equation*
17. Heiko Appel, Max Planck Institute for Structure and Dynamics of Matter, Hamburg: *Ab-initio description of classical and quantized photon fields coupled to molecular systems*
18. Shane Parker, Dept. of Chemistry, U. C. Irvine: *Nonlinear properties from TDDFT: trials and tribulations, quadratic response theory*
19. Dominik Juraschek, ETH Zurich, Switzerland: *Dynamical Multiferroicity*
20. Mark Casida, Dept. of Chemistry, University of Grenoble, France: *On the calculation of Δ⟨ S2⟩ for electronic excitations in TDDFT*
21. Xiaosong Li, Dept of Chemistry, University of Washington: *Two-Component Noncollinear Time-Dependent Density Functional Theory for Excited State Calculations*
22. Alberto Castro, Institute for Biocomputation and Physics of Complex Systems,University of Zaragoza, Spain: *Brief reports on (1) Control for quantum optics processes; and (2) Propagators for the time-dependent Kohn-Sham equations*
23. Yoshiyuki Miyamoto, National Institute of Advanced Industrial Science & Technology (AIST), Tsukuba, Japan: *Laser-field enhancement and coherent electron dynamics in low-dimensional materials examined by real-time TDDFT*
24. Dillon Yost, Univ. North Carolina, Chapel Hill: *Recent progress in modeling electronic stopping using non-equilibrium RT-TDDFT simulations*
25. Barry Dunietz, Dept of Chemistry, Kent State University: *First-principles based studies of photo-induced electron transfer and transport through molecular-resolved interfaces*
26. Michele Pavanello, Dept. of Chemistry, Rutgers-Newark: *Electronic properties of liquids and materials from subsystem and orbital-free DFT*
27. Lucia Reining, Ecole Polytechnique, Palaiseau, France: *Density functionals for the dynamic structure factor and more*
28. Kazuhiro Yabana, Center for Computational Sciences, University of Tsukuba: *Real-time TDDFT simulations for interactions of ultrashort laser pulses with solids, coupling to Maxwell's equations*
29. Norah Hoffmann, Max Planck Institute for Structure and Dynamics of Matter, Hamburg: *Ab-Initio Description of Photoinduced Processes Beyond Classical Maxwell Theory*
30. Simone Marocchi, Institute of Physics, University of Sao Paolo, Brazil: *Quantum mechanics in metric space: distances between exchange-only correlations*
31. Arindam Chakraborty, Dept of Chemistry, Syracuse University: *Linked-cluster formulation of screened electron-hole interaction from explicitly-correlated geminal functions without using unoccupied states*
32. Normand Modine, Center for Integrated Nanotechnologies, Sandia National Lab: *Long-time limit of TDDFT*
33. Cheng-Wei Lee, University of Illinois at Urbana Champaign: *Electron-Ion Dynamics in Semiconductors with defects under Ion Irradiation*

34. Neepa Maitra, Dept of Physics, Hunter College and Graduate Center CUNY: *New Approaches to Non-Adiabatic Functionals*

Of the 34 workshop speakers, 8 were students/post-docs (Gossel, Mandal, Parker, Marocchi, Lee, Hoffmann, Yost, Juraschek, Eich), 10 were early career faculty/scientists (Isborn, Narang, Schleife, Suzuki, Eich, Pavanello, Strubbe, Andrade-Valencia, Parkhill, Baczewski), and 7 were women (Mandal, Hoffmann, Maitra, Isborn, Reining, D'Amico, Narang).

Summary of Workshop Expenditures

The DOE funds from this grant ($10.5k) provided support for 33 students to attend the TSRC TDDFT summer school. This, together with MolSSI funds ($10k), covered all of the student lodging for the week of the summer school ($540 per student), and a portion of the TSRC registration costs ($45 per student). The remaining funds were used to partially support registration reimbursement for the students who stayed the week after the summer school to attend the TSRC workshop on excited states.

To summarize other support we had for the school: Additional MolSSI funds were used towards one catered working lunch in which the students set up their computing environments for the NWChem and Octopus programs ($946). RCSA funds were used to help support the travel and lodging costs of the summer school instructors from the US and for Giovanni Vignale (Missouri) who was to join for the panel discussion. Psi-k contributed support for the school instructors from Europe.