

# Robust PEXSI and fast rt-TDDFT Method

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# Outline

- ▶ Part I: Robust PEXSI method
  - ▶ Introduction
  - ▶ Robust determination of chemical potential
    - ▶ Coarse level: inertia counting
    - ▶ Fine level: multi-point evaluation of Fermi Operator Expansion
  - ▶ Parallelization
  - ▶ Numerical results
  - ▶ Conclusion
- ▶ Part II: Fast rt-TDDFT with parallel transport gauge
  - ▶ Introduction to TDDFT
  - ▶ Implicit time integrator
  - ▶ Testing results
  - ▶ Conclusion and Future work

# Fermi Operator Expansion I

FOE method expands  $\Gamma$  using a rational approximation as

$$\Gamma \approx \sum_{l=1}^P \Im (\omega_l^p (H - (z_l + \mu)S)^{-1}). \quad (1)$$

The chemical potential  $\mu$  chosen to ensure that

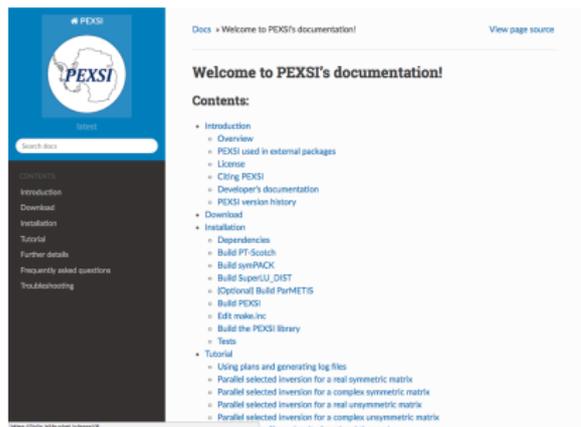
$$N_\beta(\mu) = \text{Tr}[f_\beta(\Lambda - \mu I)] = \text{Tr}[S\Gamma] = N_e, \quad (2)$$

- ▶ Computational complexity 1D system:  $O(N_e)$ ; 2D system:  $O(N_e^{1.5})$ , 3D system  $O(N_e^2)$ .
- ▶ Contour integral poles for DM, EDM and FDM.
- ▶ Easy parallelization: intra-pole and inner-pole.
- ▶ Works for both metallic system and semi-conductors.

# PEXSI: Pole EXpansion Selected Inversion package

- ▶ Before v1.0, PEXSI use 40-80 contour integral poles in real calculations
- ▶ Scale up to thousands of CPU cores for single pole
- ▶ Tow level parallelization: embarrassing parallel over the poles and  $Np_{row} \times Np_{column}$  parallel within single pole
- ▶ Integrated into BigDFT, CP2K, SIESTA, DGDFT, FHI-aims, QuantumWise ATK, ESL Bundle
- ▶ Typically used for accelerating material simulation with more than 10,000 atoms
- ▶ one challenge is determine the chemical potential  $\mu$ , so that  $|N_{e_{computed}} - N_{e_{exact}}|$  within given tolerance.

# PEXSI Version 1.0.3

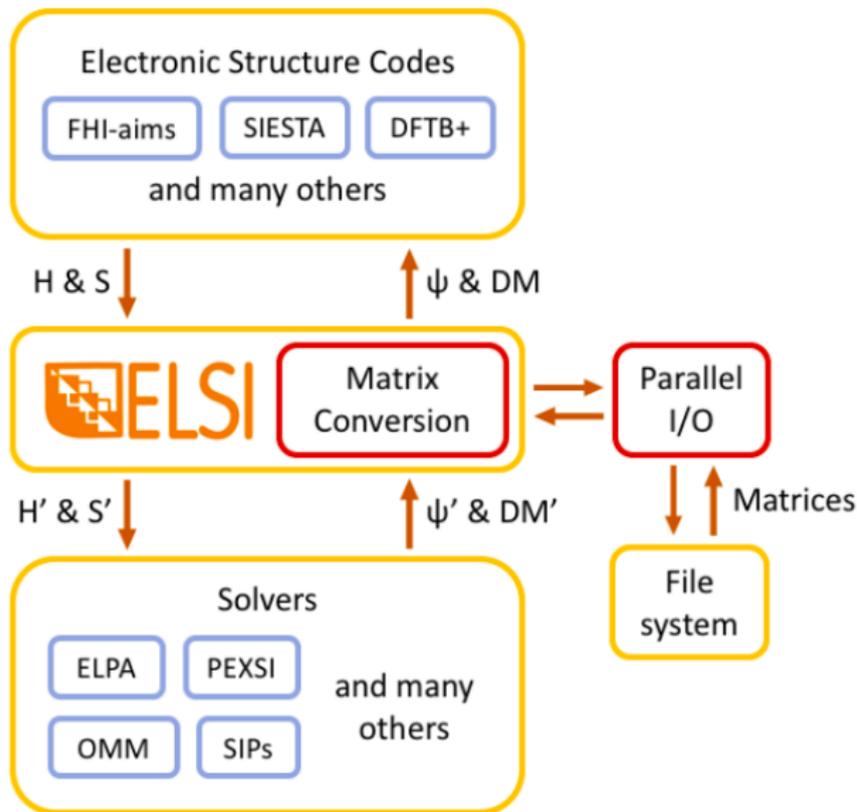


The screenshot shows the PEXSI documentation website. At the top left is the PEXSI logo, which consists of a globe with the word "PEXSI" overlaid. Below the logo is a search bar with the text "Search docs". To the left of the main content is a dark navigation menu with the following items: Introduction, Download, Installation, Tutorial, Further details, Frequently asked questions, and Troubleshooting. The main content area has a blue header with the text "Welcome to PEXSI's documentation!" and a "View page source" link. Below the header is a "Contents:" section with a list of links:

- Introduction
  - Overview
  - PEXSI used in external packages
  - License
  - Citing PEXSI
  - Developer's documentation
  - PEXSI version history
- Download
- Installation
  - Dependencies
  - Build PT-Scotch
  - Build symPACK
  - Build SuperLU\_DIST
  - Optional Build ParMETIS
  - Build PEXSI
  - Build make.inc
  - Build the PEXSI library
  - Tests
- Tutorial
  - Using plans and generating log files
  - Parallel selected inversion for a real symmetric matrix
  - Parallel selected inversion for a complex symmetric matrix
  - Parallel selected inversion for a real unsymmetric matrix
  - Parallel selected inversion for a complex unsymmetric matrix

- ▶ url: [www.pexsi.org](http://www.pexsi.org)
- ▶ includes Moussa's optimized pole expansion(DM only)
- ▶ robust chemical potential determination.

# PEXSI: part of ELSI



# Challenge for FOE methods: Determine chemical potential

The chemical potential  $\mu$  ensures:

$$N_{\beta}(\mu) = \text{Tr}[f_{\beta}(\Lambda - \mu I)] = \text{Tr}[S\Gamma] = N_e, \quad (3)$$

where  $N_e$  is the number of electrons.

**Problem:** pole expansion does not calculate eigenvalue, thus can not directly get chemical potential.

- ▶ bisection: several PEXSI evaluation FOE in single SCF.
- ▶ Newton's method: not robust, over-correction in the process.
- ▶ inertia counting + Newton's method

**our goal:** a robust and efficient chemical potential determination method

# Idea: take a look back at iterative method

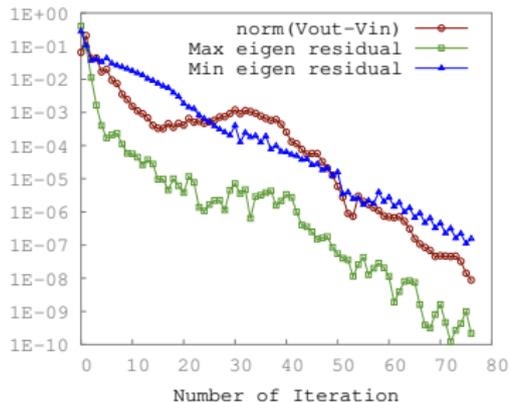
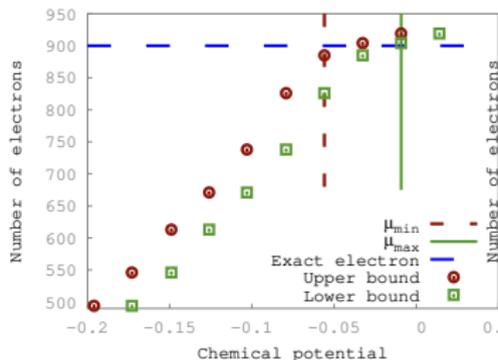


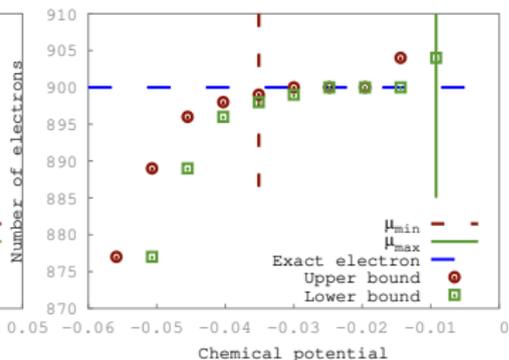
Figure 1: Si-32 system, 64 bands, PBE calculation with plane wave iterative method

- ▶ Not necessary to converge to correct chemical potential in each SCF step.
- ▶ Keep rigorous upper and lower bounds of the chemical potential
- ▶ The bounds will converge with the SCF iterations.
- ▶ Only one PEXSI evaluation will be carried in each SCF step.

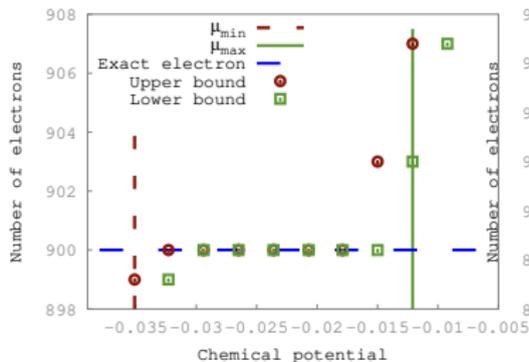
# Bounds estimation with inertia counting



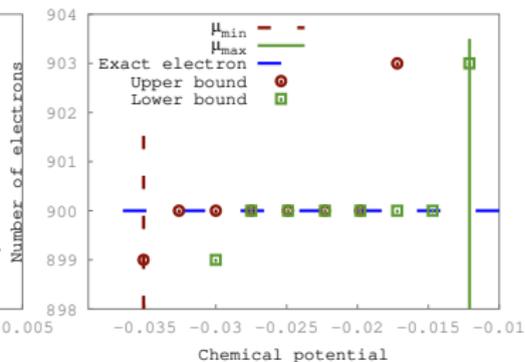
(a) step 1.



(b) step 2.



(c) step 3.



(d) step 4.

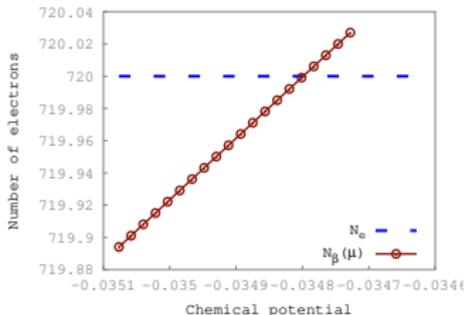
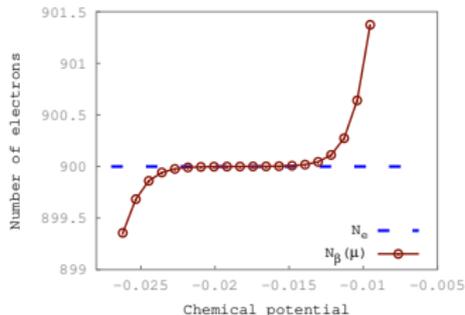
# Fine level: multiple $\mu$ evaluation of Fermi Operator

In a single SCF step:

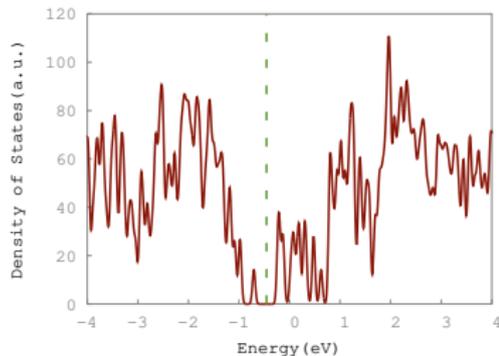
- ▶ launch  $N_\mu$  Fermi operator evaluation simultaneously.
- ▶ update the  $(\mu_{min}, \mu_{max})$  with calculated results.
- ▶ Interpolate  $(\mu_{min}, \mu_{max})$  and the DM

between SCF steps:

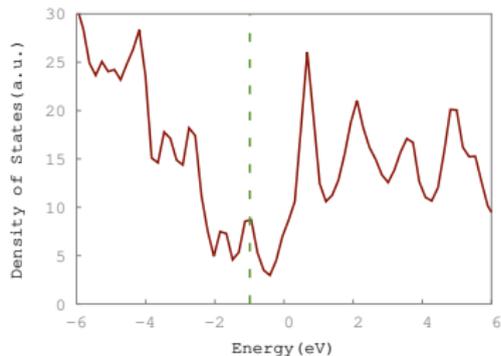
- ▶ keep rigorous bounds of  $(\mu_{min}, \mu_{max})$  by updating with  $(\mu_{min} + \Delta V_{min}, \mu_{max} + \Delta V_{max})$ .



# DOS of the PNR and GRN 180 atoms system



(a) PNR 180

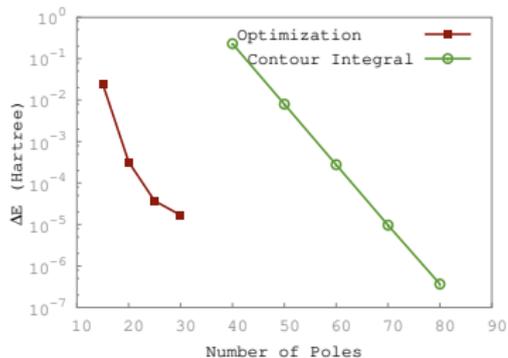


(b) GRN 180

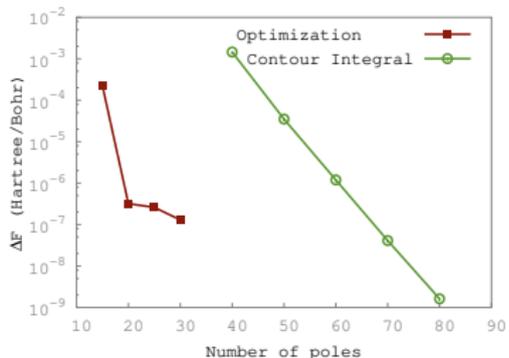
**Figure 4:** The total densities of states(DOS) of PNR and GRN 180 atoms, respectively. The fermi levels are marked by the green dash line.

The tests are performed on Edison, NERSC, with our DGDFT package.

# Pole Expansion Accuracy: Moussa's optimized and Contour integral pole expansion



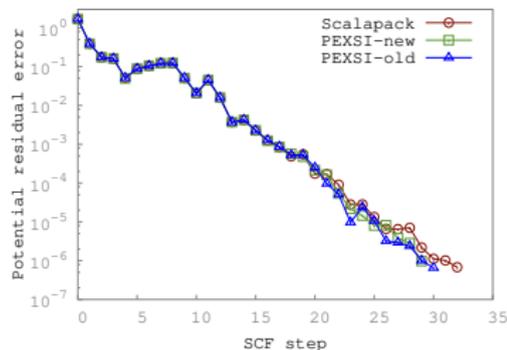
(a) Energy



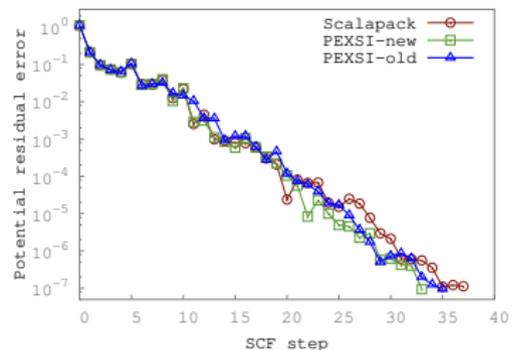
(b) Force

Figure 5: Error of the energy and force of the PEXSI method with respect to the number of poles for the graphene 180 system.

# Pole Expansion SCF Accuracy: PNR and GRN 180 system



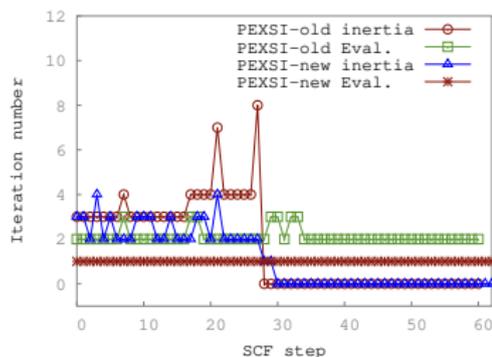
(a) PNR 180.



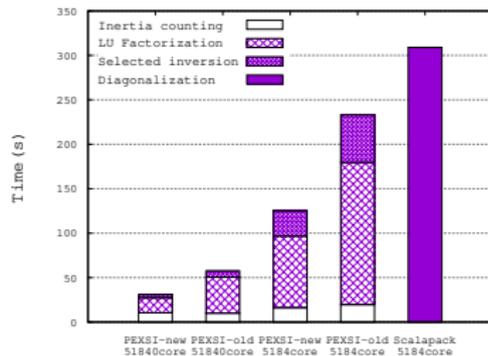
(b) GRN 180.

Figure 6: SCF convergence with ScaLAPACK and PEXSI-old and PEXSI-new along the SCF steps for PNR 180 and GRN 180 systems.

# PEXSI Timing



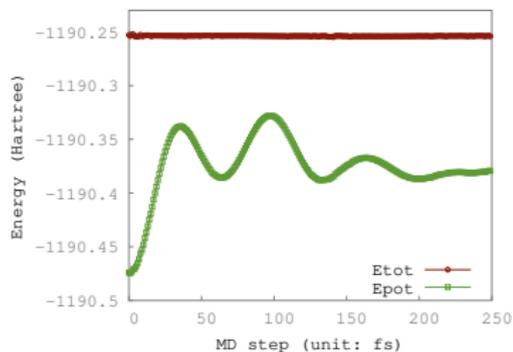
(a) Number of inertia counting step and evaluation of the Fermi operator steps.



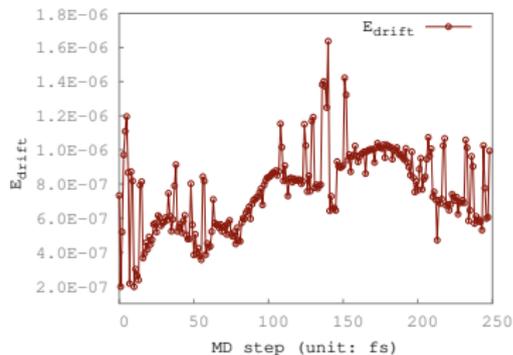
(b) The timing for PEXSI-new, PEXSI-old and diagonalization per SCF step.

Figure 7: Comparison of the performance of PEXSI-new, PEXSI-old and diagonalization for the GRN 6480 system.

# MD simulation



(a) Total and potential energy



(b) Energy drift

Figure 8: The potential energy, total energy and the drift of the total energy using the PEXSI-new method.

## Conclusion and on-going work

- ▶ We present a robust and efficient algorithm in determine the chemical potential in the PEXSI for solving KSDFT.
- ▶ Only one PEXSI evaluation is performed in a single SCF step.
- ▶ The chemical potential converges with the SCF iterations.
- ▶ Moussa's optimized pole expansion is set as default in PEXSI 1.0.3
- ▶ Two  $\mu$  parallelization is recommended for PEXSI 1.0.3
- ▶ Tests show that we have a speedup of 2x for Graphene 6480 atoms.
- ▶ The Journal of Chemical Physics 147, 144107 (2017); <https://doi.org/10.1063/1.5000255>
- ▶ on-going: optimized poles for DM, EDM, FDM in the next version.

## Part II: fast rt-TDDFT method

- ▶ Explicit time integrator.
- ▶ Parallel transport gauge and implicit time integrator
- ▶ Testing results
- ▶ Conclusion

# Time Dependent Density Functional Theory (TDDFT)

## Introduction

- ▶ Explicit time propagator, solve the TDDFT equation by directly calculate:

$$i\partial_t\psi_i(t) = H(t)\psi_i(t). \quad (4)$$

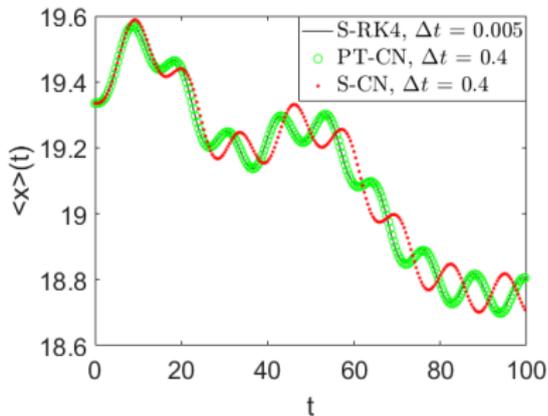
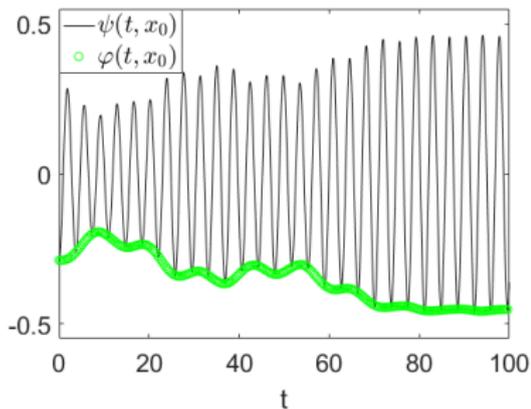
- ▶ major drawback: it requires a small time step size satisfying  $\Delta t \lesssim \|H\|^{-1}$  due to the stability restriction. For plane wave basis set, whose  $H$  spectrum is more than 40 Ryb,  $\Delta t$  is set to be smaller than 1 as.

## Implicit time propagator for TDDFT

- ▶ wavefunction  $\psi(t)$  oscillate fast, while the density matrix  $P(t)$  oscillate much slower
- ▶ An optimal gauge choice given by parallel transport formulation can approximate the orbitals into a straight line, give us a much bigger time step(50 – 100 as) in than explicit method.
- ▶ This PT-CN method can be used in any RT-TDDFT calculation package.

# Implicit time propagator for TDDFT

Real parts of  $\psi(x_0, t)$ ,  $\varphi(x_0, t)$  at  $x_0 = 25.0$  and Dipole moment



# TDDFT optimal gauge I

TDDFT equations

$$i\partial_t\psi_i(t) = H(t, P(t))\psi_i, \quad i = 1, \dots, N_e. \quad (5)$$

Here  $\Psi(t) = [\psi_1, \dots, \psi_{N_e}]$  are the electron orbitals, and density matrix  $P(t) = \Psi(t)\Psi^*(t)$ .

Suppose another set of Orbitals  $\Phi(t) = \Psi(t)U(t)$  satisfies:

$$P(t) = \Psi(t)\Psi^*(t) = \Phi(t)\Phi^*(t) \quad (6)$$

and von Neumann equation:

$$i\partial_t P = [H, P] = HP - PH. \quad (7)$$

Our goal: optimize the gauge matrix, so that the transformed orbitals  $\Phi(t)$  vary *as slowly as possible*, without altering the density matrix. This results in the following variational problem

$$\min_{U(t)} \|\dot{\Phi}\|_F^2, \text{ s.t. } \Phi(t) = \Psi(t)U(t), U^*(t)U(t) = I_{N_e}. \quad (8)$$

## TDDFT optimal gauge II

Here  $\|\dot{\Phi}\|_F^2 := \text{Tr}[\dot{\Phi}^* \dot{\Phi}]$  measures the Frobenius norm of the time derivative of the transformed orbitals. The minimizer of (8), in terms of  $\Phi$ , satisfies

$$P\dot{\Phi} = 0. \quad (9)$$

implicitly defines a gauge choice for each  $U(t)$ , and this gauge is called the *parallel transport gauge*. The governing equation of  $\Phi$ , together with the von Neumann equation, can be written as

$$i\partial_t \Phi = H\Phi - \Phi(\Phi^* H\Phi), \quad P(t) = \Phi(t)\Phi^*(t). \quad (10)$$

Preprint: Fast real-time time-dependent density functional theory calculations with the parallel transport gauge

## PT-CN method: implicit time integrator I

The implicit Crank-Nicolson scheme for the parallel transport dynamics (PT-CN):

$$\begin{aligned} & \Phi_{n+1} + i \frac{\Delta t}{2} \{ H_{n+1} \Phi_{n+1} - \Phi_{n+1} (\Phi_{n+1}^* H_{n+1} \Phi_{n+1}) \} \\ & = \Phi_n - i \frac{\Delta t}{2} \{ H_n \Phi_n - \Phi_n (\Phi_n^* H_n \Phi_n) \}. \end{aligned} \tag{11}$$

The equation can be solved self-consistently with a preconditioned Anderson mixing scheme.

$\Delta t = 10 - 100$  as.

# TDDFT tests compare between S-RK4 and PT-CN

- ▶ TDDFT with an external field.
  - ▶ optical spectrum of anthracene
  - ▶ place benzene in a laser field
- ▶ Ultra-fast ion collision
  - ▶ fast Cl<sup>-</sup> ion collide into a 112 atom Graphene sheet

# Anthracene optical spectrum

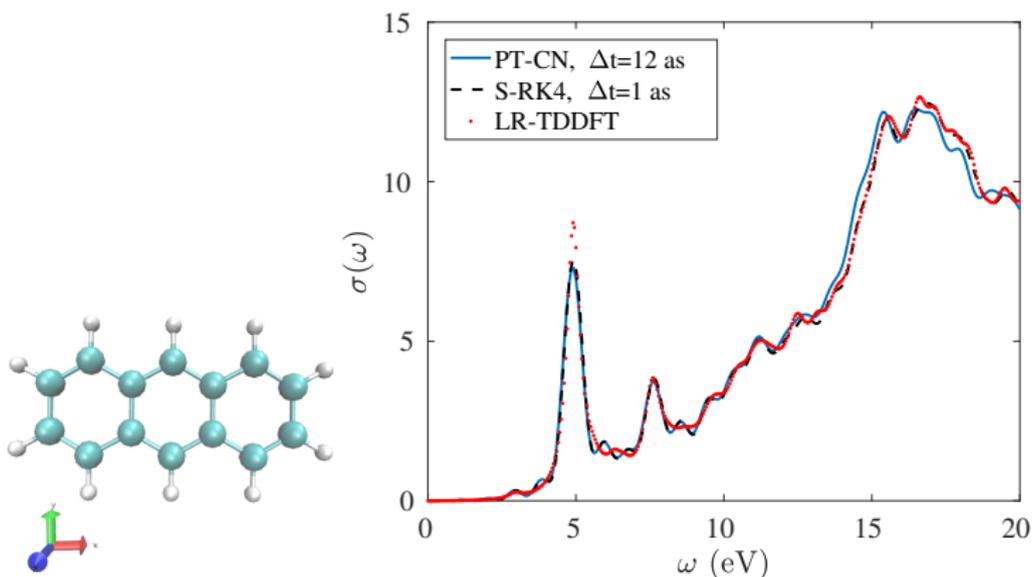
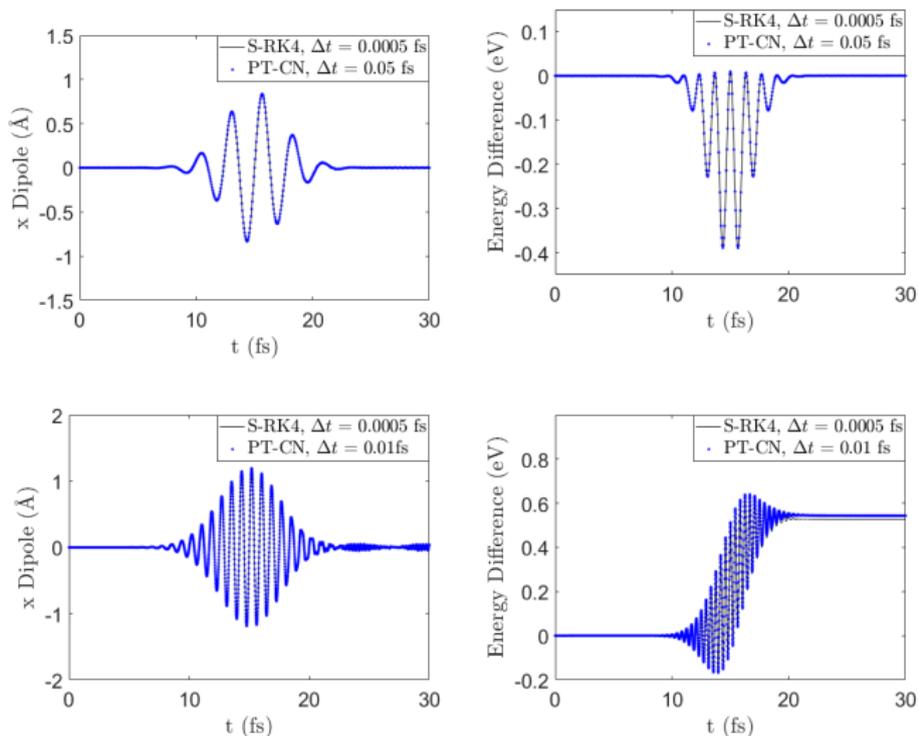


Figure 9: Anthracene and its absorption spectrum.

# Benzene driven by a laser external field



**Figure 10:** Electronic dynamics of a benzene driven by a laser with wavelength  $\lambda = 800$  nm in (a)(b), 250 nm in (c)(d).

## Benzene with laser – accuracy and efficiency

Method	$\Delta t$ (fs)	AEI (eV)	AOE (eV)	MVM	Speedup
S-RK4	0.0005	0.5260	/	152000	/
PT-CN	0.005	0.5340	0.0080	28610	5.3
PT-CN	0.0065	0.5347	0.0087	22649	6.7
PT-CN	0.0075	0.5362	0.0102	21943	6.9
PT-CN	0.01	0.5435	0.0175	15817	9.6
PT-CN	0.02	0.5932	0.0672	12110	12.6

**Table 1:** The accuracy is measured using the average energy increase (AEI) after 25.0 fs and the average overestimated energy (AOE) after 25.0 fs. The efficiency is measured using the total number of matrix-vector multiplications (MVM) during the time interval from 5.5 fs to 24.5 fs, and the computational speedup.

# Cl- ion collide with graphene sheet

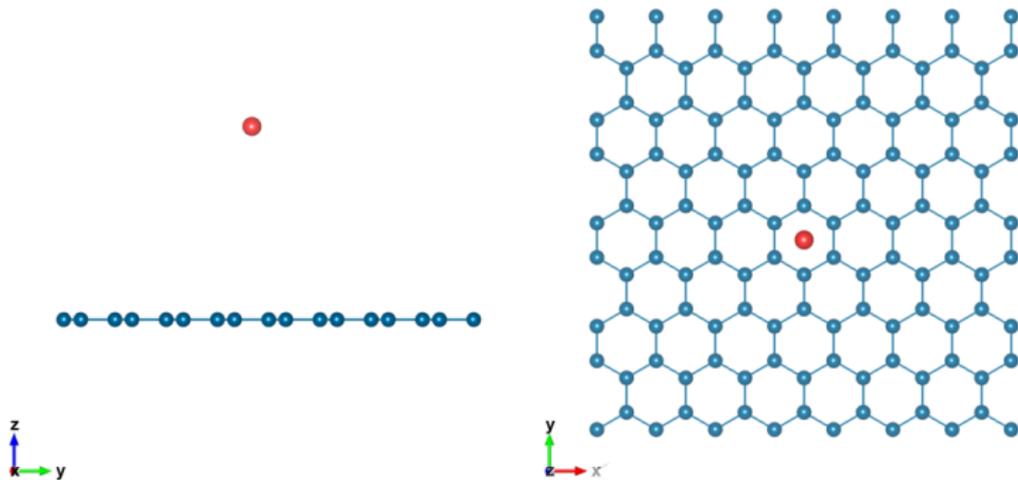
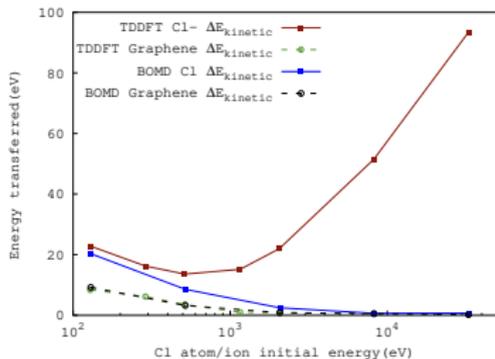
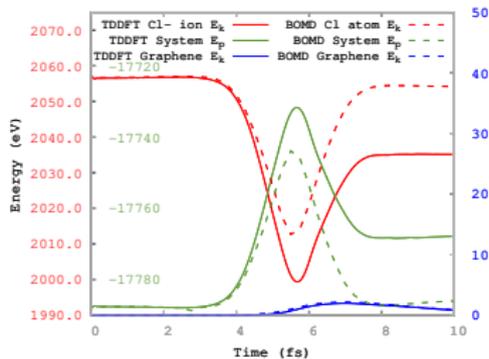


Figure 11: Model of Cl-Graphene collision. Red point is the center of the graphene ring.

# BOMD & TDDFT – Ion collision



(a) BO-MD and TDDFT energy transfer with different initial kinetic energies.



(b) BO-MD and TDDFT energy transfer with time, projectile speed is 2.0bohr/fs.

Figure 12: Energy transfer comparison between BOMD and TDDFT.

## Density of States – Ion collision

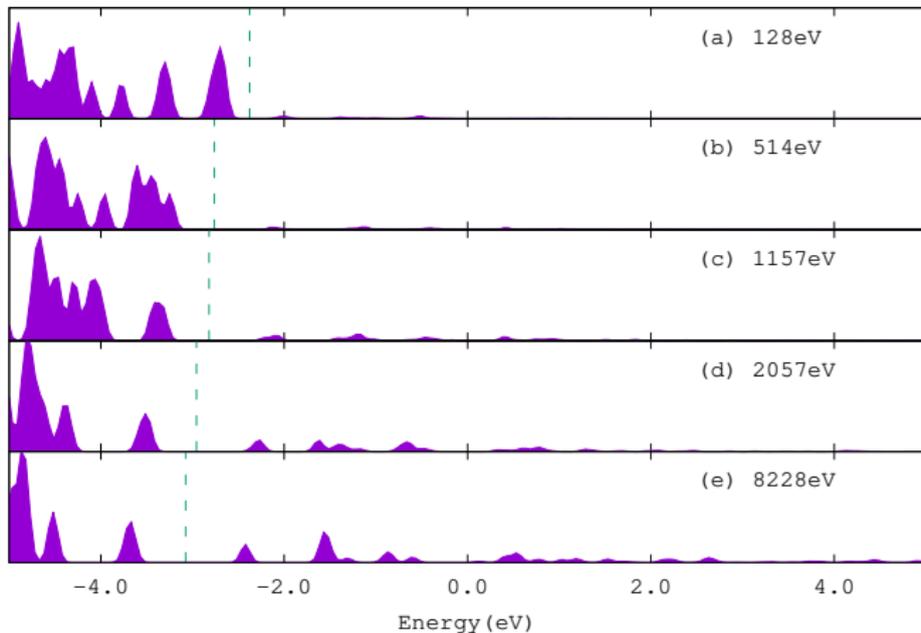


Figure 13: Density of state after the ion collision. Green dashed line: Fermi energy.

# Currently working on: Hybrid functional TDDFT

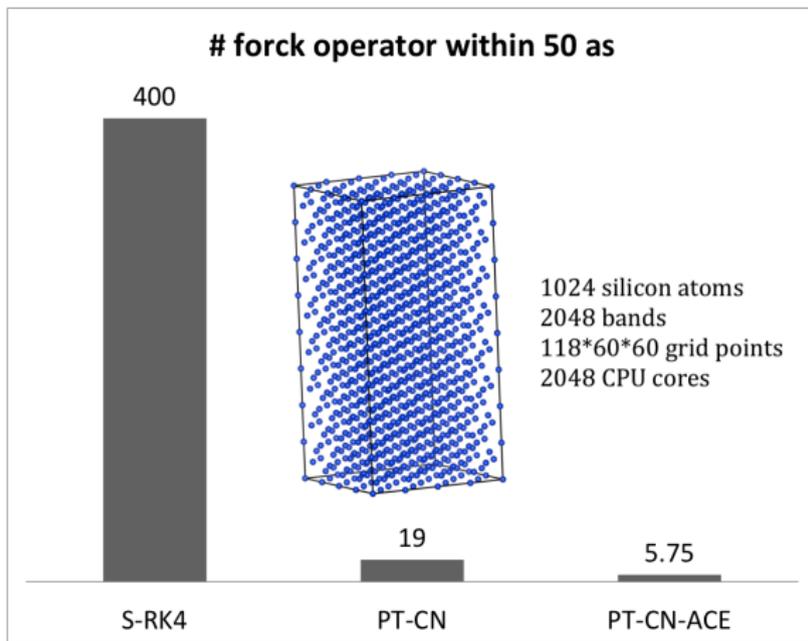


Figure 14: Plane wave TDDFT with 1024 atoms under a 389 nm field

Paper in preparation.

## Conclusion & on-going work

- ▶ An optimized gauge choice to improve the numerical efficiency with no loss of accuracy is demonstrated.
- ▶ Tests show it can speedup the RT-TDDFT by a factor of 5-10x for systems between 32-1000 atoms.
- ▶ GPU hybrid TDDFT method next.

Thank you for your attention



Acknowledgement

