DGDFT: A Massively Parallel Method for Large Scale Density Functional Theory Calculations

Wei Hu,[1,*] Lin Lin,[2,1,*] Chao Yang,[1,*] Gaigong Zhang,[1] Mathias Jacquelin,[1] Eugene Vecharynski,[1] and John E. Pask[3,*]

Email: whu@lbl.gov; linlin@math.berkeley.edu; cyang@lbl.gov; pask1@llnl.gov

[1]Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[2]Department of Mathematics, University of California, Berkeley, CA 94720, USA

[3]Physics Division, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

DGDFT Collaboration

Support provided through the SciDAC program funded by U.S. Department of Energy, Office of Science, Advanced Scientific Computing Research and Basic Energy Sciences.

Goals

DGDFT: Discontinuous Galerkin Method for Density Functional Theory

http://www.dgdft-scidac.org/

A new method and code for quantum molecular dynamics (QMD) simulations, capable of modeling complex, mixed-phase systems, containing 10,000 atoms or more.

Apply the new methodology to understand the detailed chemistry and dynamics of the solid-electrolyte interphase (SEI) layer in Li-ion cells, a key factor in performance, lifetime, and safety.

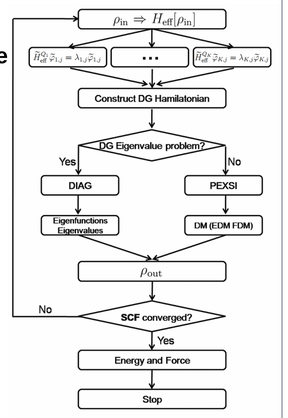
Theory

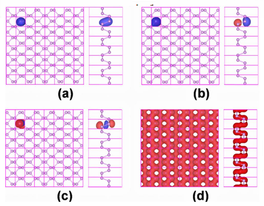
Adaptive Local Basis (ALB) Set

Discontinuous, local, systematically improvable

Effect of chemical environment

$$E_{\mathrm{DG}}(\{\psi_i\}) = \frac{1}{2}\sum_{i=1}^{N}\langle\nabla\psi_i,\nabla\psi_i\rangle_{\mathcal{T}} + \langle V_{\mathrm{eff}},\rho\rangle_{\mathcal{T}}$$
$$+\sum_{I=1}^{N_A}\sum_{\ell=1}^{L_I}\gamma_{I,\ell}\sum_{i=1}^{N}\left|\langle b_{I,\ell}(\cdot-R_I),\psi_i\rangle_{\mathcal{T}}\right|^2$$
$$-\sum_{i=1}^{N}\langle\{\{\nabla\psi_i\}\},[[\psi_i]]\rangle_{\mathcal{S}} + \alpha\sum_{i=1}^{N}\langle[[\psi_i]],[[\psi_i]]\rangle_{\mathcal{S}}$$





For 2D phosphorene P_{140}

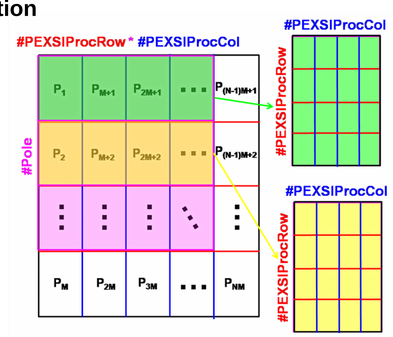
1 × 8 × 8 2D element partition

Pole Expansion and Selected Inversion (PEXSI)

Fermi operator expansion http://www.pexsi.org/

Solving KSDFT without diagonalization

$$\rho \approx \mathrm{diag}\sum_{i=1}^{Q}\frac{\omega_i}{H - z_i I}$$



Two Level Parallelization

Highly scalable up to 128,000 cores

***Intra-element* parallelization**

numBasis: 10 ~ 100 cores

***Inter-element* parallelization**

numElement: 4 ~ 10,000 cores

Accuracy and efficiency

Computational Accuracy

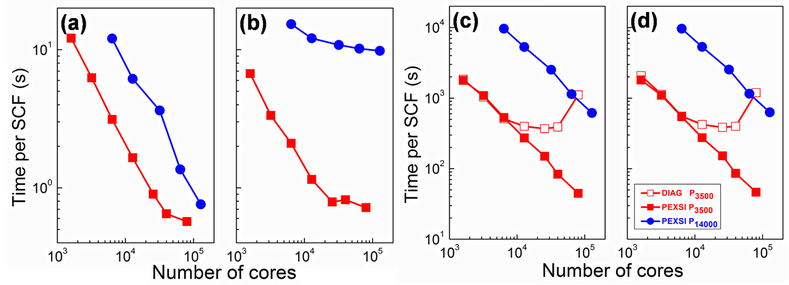
Total energy and atomic forces for 2D phosphorene

DGDFT P_{140}		DIAG		PEXSI	
Ecut	#ALB	ΔE	ΔF	ΔE	ΔF
20	91.43	5.22E-04	4.03E-03	3.22E-04	4.03E-03
40	18.28	4.51E-02	5.97E-02	4.57E-02	5.97E-02
40	27.42	6.67E-04	2.51E-03	6.85E-04	2.52E-03
40	36.57	1.34E-04	6.16E-04	1.59E-04	6.18E-04
40	45.71	7.00E-05	4.00E-04	6.44E-05	5.23E-04
40	91.43	-4.32E-07	5.93E-04	1.34E-04	5.93E-04
100	91.43	1.59E-05	1.97E-04	8.04E-05	1.97E-04
200	91.43	3.39E-06	1.06E-04	8.12E-05	1.06E-04

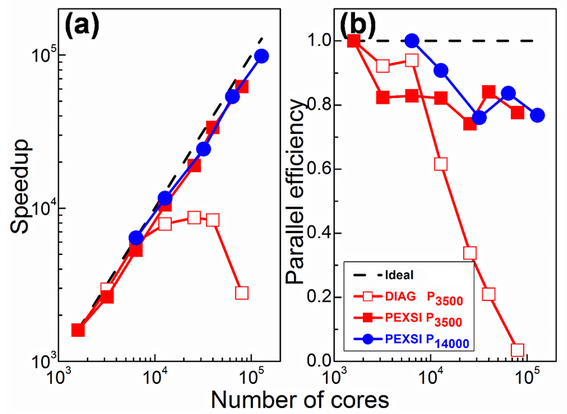
Computational Time and Parallel Efficiency

Highly scalable up to 128,000 cores (3500 and 14000 atoms)

High parallel efficiency up to 80% for 2D phosphorene



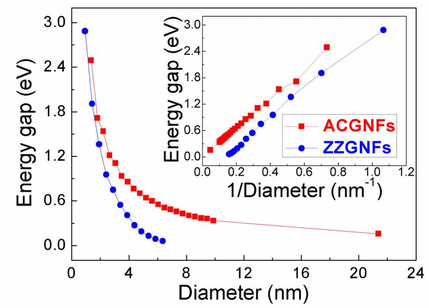
Generate ALB basis, Construct DG Hamiltonian, Diagonalize DG Hamiltonian

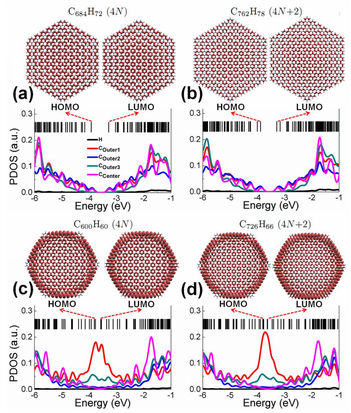


Applications

Graphene Nanoflakes

11700 atoms SIESTA-PEXSI





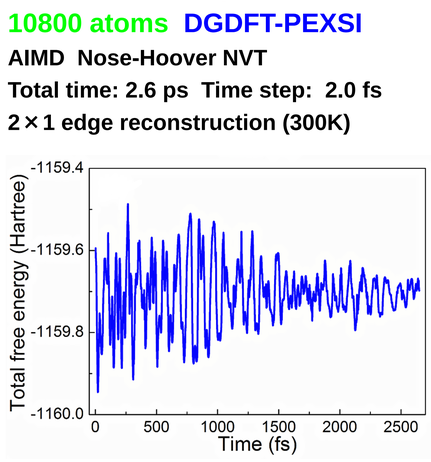
Armchair Phosphorene Nanoribbons

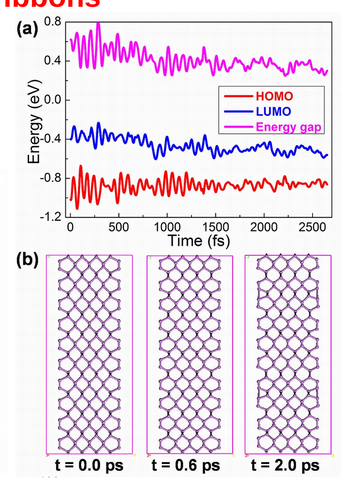
10800 atoms DGDFT-PEXSI

AIMD Nose-Hoover NVT

Total time: 2.6 ps Time step: 2.0 fs

2 × 1 edge reconstruction (300K)





Li-ion anode-electrolyte interface

4720 atoms DGDFT-PEXSI

DGDFT:

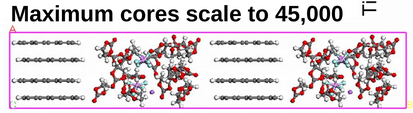
1 × 30 × 30 2D element partition

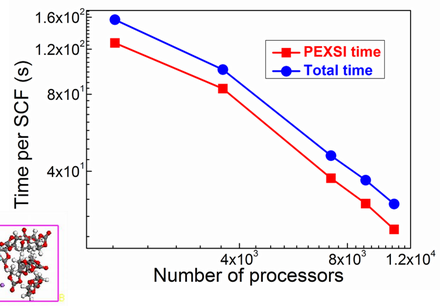
22 basis functions per atom

PEXSI:

50 poles, 30 × 30 cores per pole

Maximum cores scale to 45,000





Lin Lin, Jianfeng Lu, Lexing Ying and Weinan E, Adaptive local basis set for Kohn–Sham density functional theory in a discontinuous Galerkin framework I: Total energy calculation, J. Comput. Phys. 231, 2140 (2012).

Gaigong Zhang, Lin Lin, Wei Hu, Chao Yang and John E. Pask, Adaptive lo cal basis set for Kohn-Sham density functional theory in a discontinuous Galerkin framework I I: Force, vibration, and molecular dynamics calculations, J. Comput. Phys. In preparation, (2015).

Wei Hu, Lin Lin and Chao Yang, DGDFT: A massively parallel method for large scale density functional theory calculations, J. Chem. Phys. Submitted, (2015).

Lin Lin, Jianfeng Lu, Lexing Ying, Roberto Car and Weinan E, Fast algorithm for extracting the diagonal of the inverse matrix with application to the electronic structure analysis of metallic systems, Commun. Math. Sci. 7, 755 (2009).

Lin Lin, Mohan Chen, Chao Yang and Lixin He, Accelerating atomic orbital-based electronic structure calculation via pole expansion and selected inversion, J. Phys.: Condens, Matter 25, 295501 (2013).

Lin Lin, Alberto García, Georg Huhs and Chao Yang, SIESTA-PEXSI: Massively parallel method for efficient and accurate ab initio materials simulation without matrix diagonalization, J. Phys.: Condens. Matter 26, 305503 (2014).

Wei Hu, Lin Lin, Chao Yang and Jinlong Yang, Electronic structure and aromaticity of large-scale hexagonal graphene nanoflakes, J. Chem. Phys. 141, 214704 (2014).

Wei Hu, Lin Lin and Chao Yang, Edge eeconstruction in armchair phosphorene nanoribbonsrRevealed by discontinuous Galerkin density functional theory, Phys. Chem. Chem. Phys. DOI: 10.1039/C5CP00333D Accepted, (2015).

Wei Hu, Lin Lin, Chao Yang and Jinlong Yang, Edge-controlled phosphorene nanoflakes heterjunctions as ultrathin solar cells, In preparation, (2015).