Novel First-principle Simulations of $3D$ Atomistic Structure

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ABSTRACT

In order to address the current computational challenges posed by First-principle simulation of $3D$ devices, a high order real space mesh technique (FEM) is employed to reduce the size of the system matrix and improve the convergence. Furthermore, the CMB strategy and the FEAST algorithm have been proposed to compute the electron density efficiently. The proposed techniques can benefit the $3D$ simulations of atoms, molecules, and nanowire-type devices as well.

Keywords: First-principle calculation, molecule simulations, electron density calculation, finite element method.

1 Introduction

Modeling approaches in computational electronics have become increasingly driven by the need of more fundamental and comprehensive understanding of the nanoscale physics of systems only through knowledge of the constituent atoms. First principle modeling, which starts directly at the level of established laws of physics and does not include any fitting parameters, provides both reliability and flexibility to obtaining the electronic properties of arbitrary nanostructures and devices. In this paper, Density Functional Theory(DFT), one of first principle methods, is employed for performing electronic structure calculations, and the self-consistent Kohn-Sham procedure outlined below[1] provides a practical approach for applying the DFT:

$$\left(-\frac{1}{2m}\nabla^2 + V_{eff}[n(r)]\right)\psi_i(r) = E_i\psi_i(r), \quad (1)$$

$$n(r) = 2\sum_{i=1}^{N_E}|\psi_i(r)|^2. \quad (2)$$

$$V_{eff}[n(r)] = V_{ion}(r) + V_H[n(r)] + V_{XC}[n(r)], \quad (3)$$

$$-\nabla^2 V_H(r) = \frac{\rho(r)}{\epsilon}. \quad (4)$$

$$V_{XC}[n](r) = \frac{\delta}{\delta n(r)}E_{XC}[n] \quad (5)$$

This self-consistent DFT/Kohn-Sham procedure leads to the eigenvalue Schrödinger-type equation (1) whose solution yields the orthonormal eigenstates ψ_i, which in turn, reproduce the density of the original many-body system (2). V_{eff} (3) is the effective single-particle potential, which includes V_{ion} the core potential (or ab-initio pseudopotential), V_{XC} the exchange-correlation potential, and V_H (the Hartree term), the solution of the Poisson equation (4), in which $\rho(r)$ is the charge density. Though the exact form of V_{XC} is not known, many approximations are available such as the local density approximations (LDA), in which exchange-correlation (XC) energy functional depends solely upon the value of the electronic density at each point in space.

2 Numerical Procedure

In order to perform these numerical calculations, the physical models are first discretized by expanding the wave functions over a set of basis functions [2]. In our approach, we consider the use of real-space mesh techniques such as finite element methods (FEM) (as an alternative to other widely used techniques e.g. LCAO and plane waves) for addressing the current challenges in large-scale atomistic simulations of materials and devices [3], [4]. Considering V_{ion} in (3), it could either be hard core potential or ab-initio pseudopotential. An appropriate option has then been proposed to preserve the local property of the potential in the fundamental description of the electronic structure using hard-core potential. However, it is difficult to capture the deep variation of potential around the atoms using first order FEM basis functions. Even though the linear basis is easiest to implement, for a smooth solution the difference in the accuracies between the linear and higher-order element calculations is quite important; therefore, it is necessary to implement high-order elements in order to achieve a better convergence. In our work, The third order FEM basis functions, which are derived using Legendre polynomials, and fine local meshes around the nuclei are employed to capture the deep core potential and improve the convergence properties of our all-electron calculations approach.

3 Techniques of Calculating Electron Density

Becuase of the high computational cost of calculating the electron density (2) at each step, calculations have been limited to small systems involving only few atoms. However, The number of atoms contained in nanostructures usually ranges from few hundreds to few millions; therefore, for large systems, it poses a unique challenge for electronic structure and transport calculations. In order to address the high numerical cost for computing the electron density of large-scale atomistic nanowire devices, we have investigated the mode decomposition techniques (i.e. mode approach) for solving the Schrödinger-type equation within a real-space mesh framework. In [5], compared the technique to full $3D$ discretization, it is shown that the mode approach within a real-space mesh discretization framework, can drastically reduce the numerical complexity of 3D nanowire problems and naturally leads to narrow banded system matrices [6]. The main idea is to reduce the dimension of the 3D Schrödinger-type equation by forming a basis set in some directions of the quantum confinement. There are two versions of mode decomposition (1) full mode approach (2) asymptotic mode approach. The full mode approach is indeed more robust than its asymptotic version and fewer modes are usually needed to capture the physics. However, it presents higher numerical costs in the preprocessing stage since multiple $2D$ eigenvalue problems are needed to be solved along the longitudinal direction (in contrast to one unique $2D$ eigenvalue problem for the asymptotic version). Furthermore, the mode approach can be of benefit to two distinct highly efficient numerical procedures for computing the electron density:(1) CMB strategy (2) FEAST algorithm

3.1 CMB Strategy

The CMB approach (Figure 1), which is fundamentally designed for parallel implementation, has shown robustness and numerical linear scaling performances for computing the electron density of CNT devices for a given potential. The simulation time for computing the electron density of the 48-unit (13,0) CNT, $20nm$ long and composed by 2498 atoms, is 90 seconds for a given atomistic potential on current workstation (see Figure 2).

3.2 FEAST Algorithm

Although electron density can be computed by CMB without solving the traditional eigenvalue problem, in some applications such as bandstructure calculations, obtaining explicitly the eigenvalues and eigenvectors can be of prime importance. FEAST, proposed in [7], is a general eigenvalue solver that can be used for this purpose. Its main computational tasks consist of solving few (standard) independent linear systems with multiple right-hand sides along the complex contour. Table 1 summarizes the timings obtained for computing the electron density of a (5,5) CNT using the CMB strategy and the FEAST algorithm. Both algorithms exhibit linear scaling performances when the size of the system increases. CMB is faster than FEAST but this latter is more accurate since it provides as well all the exact eigenvalues and eigenvectors.
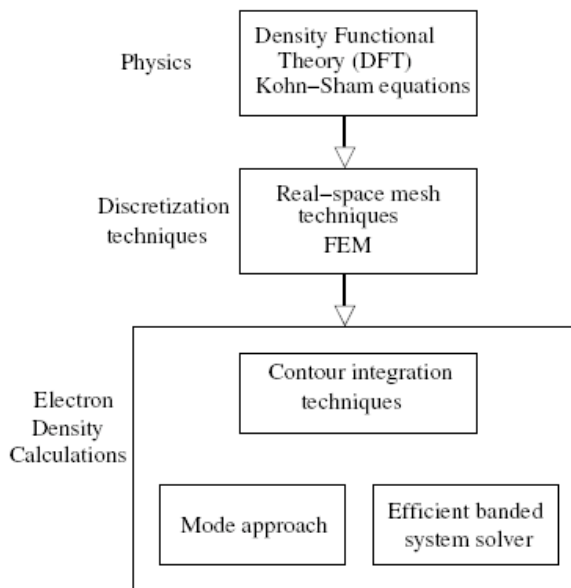

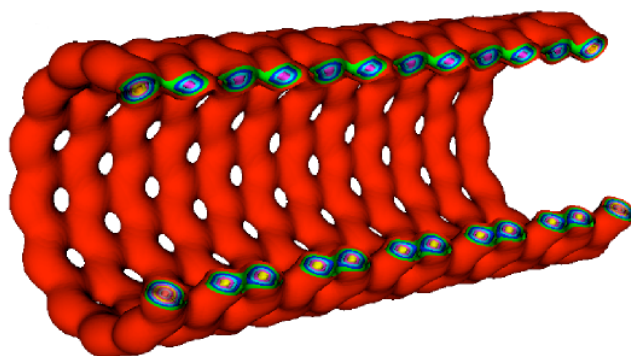Figure 1: CMB: Contour integration - Mode decomposition - banded System Solver

Figure 2: Representation of electron density for 48 unit CNT(13,0)

#unit cells	1	3	6	24	48
N	199,874	331,298	528,434	1,711,250	3,288,338
$N_1 * M$	6278	9438	14861	48125	92477
$L(nm)$	0.73	1.23	1.98	6.51	12.55
#atoms	20	60	120	480	960
$T_{CMB}(s)$	2.24	3.26	4.87	16.5	30.1
$T_{FEAST}(s)$	10.2	15.0	24.6	221	491

Table 1: Comparisons of the simulation times for computing the electron density of a metallic (5,5) CNT for a given local potential using the CMB strategy and the FEAST algorithm which computes all the eigenpairs. The resulting error on the electron density (relative residual) obtained with the two techniques, is 0.7% for the 48 unit-cells case. The calculations are performed using only one core of a Intel-Clovertown system ($2.66 Ghz$, $16 Gb$).

4   All-electron Simulation

The results discussed above are based on local empirical pseudopotential. In order to account for the information of the core electrons, we can either use an ab-initio pseudopotential or include the bare core potential in the Kohn-Sham equations. Even though the local part of ab-initio pseudopotential is smooth, the non-local part would affect the linear scalability of both CMB and FEAST. In the other hand, the core potential is local and both techniques can be taken efficiently to compute the density. In this paper, we propose to perform all electron calculations for atoms, molecules and nano-wire type devices.

4.1   Atoms, Molecules Simulation

The Figure 3 shows the mesh of molecule benzene. Because of the flexibility of FEM, fine mesh can be created around the objects of interest, and coarse mesh is generated in the regions where less points are needed. The total energy of some selected atoms and molecules calculated using the LDA Ceperley-Alder parametrization are presented in Tables 2, and this table shows satisfactory agreements with the results published in [8].

Atoms, molecules	C	O	H_2	CO	C_6H_6
Total Energy	-1018.47	-2026.75	-31.24	-3060.88	-6263.53

Table 2: Total energy (in eV) of C, and O atoms and H_2, CO, and C_6H_6 molecules calculated using high-order FEM with degree $p = 3$.

4.2   Carbon Nanotube simulation (CNT)

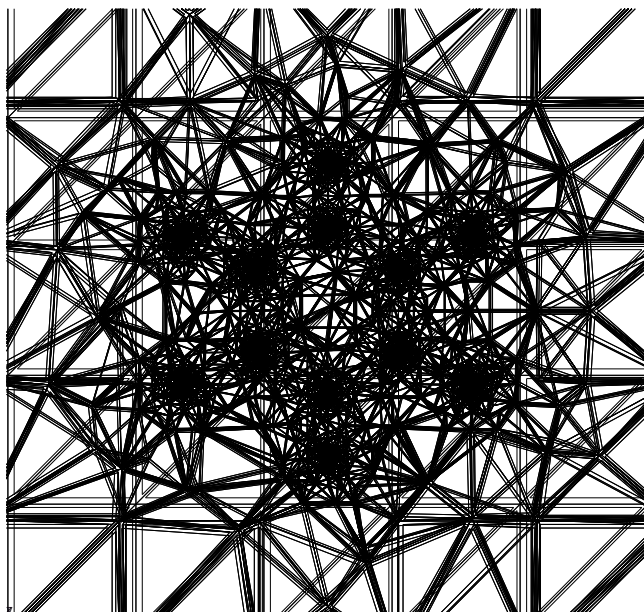For nano-wire devices, like CNT, we can adopt two different meshes:(1) prism mesh. It is generated by $2D$

Figure 3: Representation of $3D$ tetrahedral mesh of C_6H_6

$\times 1D$ mesh, and the Figure 4 shows $2D$ mesh, which is repeated along the longitudinal direction. Both CMB and FEAST can be performed on prism mesh. For all-electron self-consistent simulation, the solution converges in less than 10 iterations in 3 minutes on one core of current workstation by using CMB (Figure 6). (2)tetrahedral mesh. It is a $3D$ mesh (see Figure5). Because mode approach is based on expanding $3D$ wavefunctions using $2D$ basis, CMB is not easily amenable to tetrahedral mesh, which only works with $3D$ basis. However, it can still speed up the simulation in the sense that less elements are necessary in tetrahedral mesh to have a better approximation of wavefunctions

5   Conclusion

We have shown the simulation results of electron calculation for atoms, molecule and CNT within a real space mesh discretization framework. In order to achieve a better converge, high order FEM is employed to create a set of basis functions. The proposed CMB strategy and FEM eigenvalue solver have demonstrated scalability performance.
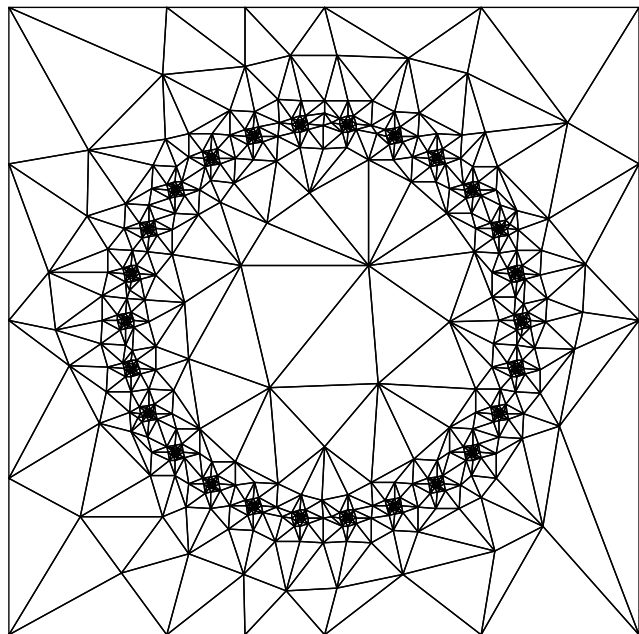
Acknowledgment

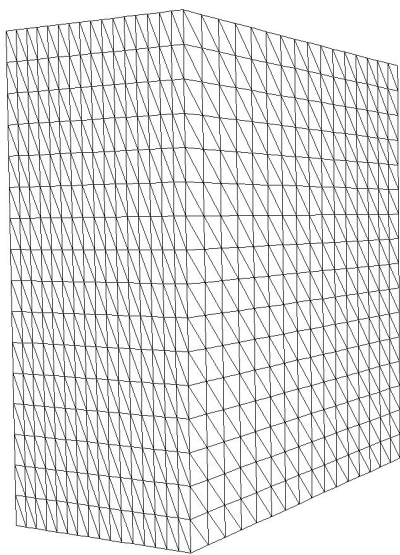Figure 4: Representation of $2D$ mesh for CNT(13,0) on closed section

Figure 5: Representation of $3D$ tetrahedral mesh for CNT(13,0).
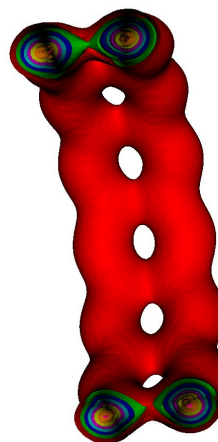

Figure 6: Representation of $3D$ electron density for CNT(13,0)

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