

Large-Scale Electronic Structure Calculations with Real-Space grid Density-Functional Theory code

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1 Introduction

First-principles quantum-mechanical electronic structure calculations based on the density-functional theory (DFT) is an important ingredient of material sciences. The system sizes handled in usual DFT calculations are limited within a thousand of atoms due to the computational complexity of fully quantum-mechanical approaches.

Real-space DFT (RSDFT) code, which has been developed in our group, is a promising application to overcome the size difficulty in first-principles quantum mechanical calculations. RSDFT is developed based on the real-space grid formulation of DFT [1] instead of conventional reciprocal-space formulation, in which frequent use of fast Fourier transform (FFT) is inevitable, so that it's suitable for massively-parallel computers such as K computer [2]. Recently, we have performed electronic structure calculations for silicon nanowires with 10,000 to 1000,000 atoms by using RSDFT on the K computer with very high performance, and we got Gordon-Bell prize in 2011 with this work.

In this paper, I review the first-principles electronic structure calculations in DFT and our program code RSDFT. In addition, I give an introduction on the technological importance of our target system silicon nanowire (SiNW), and discuss the results of the calculations for that system.

2 First-principles quantum mechanical calculations based on the density functional theory

The foundation of DFT has been given by Hohenberg and Kohn in 1964 [3], and subsequently, a formulation which is suitable for practical calculations has been given by Kohn and Sham in 1965 [4]. In the Kohn-Sham formulation of DFT, total energy of the system and the electron density are obtained by a set of functions, called the Kohn-Sham orbitals $\psi_n(\mathbf{r})$, as follows [1]:

$$\begin{aligned}
 E[\{\psi\}] = & \sum_{n=1}^N \frac{1}{2} \int d\mathbf{r} \psi_n(\mathbf{r}) \nabla^2 \psi_n(\mathbf{r}) \\
 & + \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + E_{XC}[\rho] \\
 & + \int d\mathbf{r} \rho(\mathbf{r}) v_{ION}(\mathbf{r}) \\
 & + \sum_{n=1}^N \sum_{k=1}^{N_{ION}} \int d\mathbf{r} \psi_n(\mathbf{r}) v_{PP}^k(\mathbf{r}) \int d\mathbf{r}' v_{PP}^k(\mathbf{r}') \psi_n(\mathbf{r}')
 \end{aligned} \quad (1)$$

where

$$\rho(\mathbf{r}) = \sum_{n=1}^N |\psi_n(\mathbf{r})|^2 \quad (2)$$

is the electron density, N and N_{ION} is the number of electrons and ions in the system, respectively, v_{ION} and v_{PP}^a represent the

Coulomb interaction between electrons and ions [5]. E_{XC} is called the exchange-correlation energy which arise from quantum mechanical effects. The exact functional form of the exchange-correlation energy is unknown, so that simple approximate forms, such as local-density approximation [6], are utilized for practical calculations.

Minimization of the total-energy functional with respect to the Kohn-Sham orbitals, with ortho-normalization conditions among the orbitals, leads the following equation:

$$\hat{h}_{KS} \psi_n(\mathbf{r}) = \epsilon_n \psi_n(\mathbf{r}) \quad (3)$$

$$\begin{aligned}
 \hat{h}_{KS} = & -\frac{1}{2} \nabla^2 + \int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + \frac{\delta E_{XC}[\rho]}{\delta \rho(\mathbf{r}')} \\
 & + v_{ION}(\mathbf{r}) + \sum_{k=1}^{N_{ION}} v_{PP}^k(\mathbf{r}) \int d\mathbf{r}' v_{PP}^k(\mathbf{r}')
 \end{aligned} \quad (4)$$

This is called the Kohn-Sham equation [4] which is the basic equation of first-principles electronic structure calculations. It is worse noting that Kohn-Sham equation is not a usual eigenvalue problem but nonlinear one, because the integro-differential operator Eq. (4) depends on the eigenvector itself through the electron density Eq. (2). Therefore, we have to solve the Kohn-Sham equation (nonlinear eigenvalue problem) self consistently.

3 Real-space grid DFT code

In the real-space grid calculations, we discretize the Kohn-Sham equation in 3-D spatial grid as shown in Fig. 1, and represent the differential operator in a higher-order finite-difference approximation. Then the Kohn-Sham equation is recast into a conventional linear-algebraic eigenvalue problem.

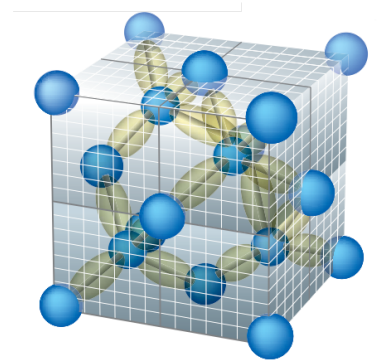


Fig. 1 Real-space grid representation of the unit cell of crystalline silicon. The eight sub cells are allocated on each CPU (or node) for parallel computations.

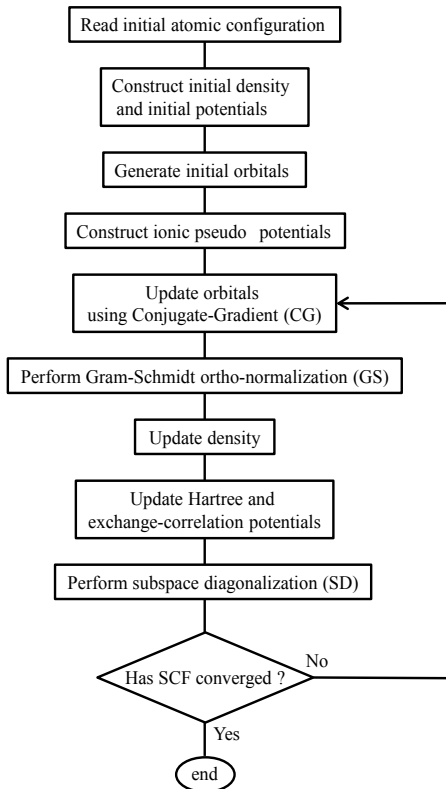


Fig. 2 Flow chart of self-consistent procedure of RSDFT calculations.

We divide the spatial region into several sub regions, and allocate the work on each region to each CPU (or node). In the finite-difference calculations, we need to exchange boundary values between next-nearest nodes, and we use *MPI_ISEND* and *MPI_IRECV* routines for this purpose. For global summations (integrations), we use *MPI_ALLREDUCE*. In addition to the grid parallelization, we also parallelize in the Kohn-Sham orbitals. In each node, we can also parallelize tasks in thread by using OpenMP.

The flow chart of the RSDFT calculations is shown in Fig. 2. The overall algorithm is the subspace-iteration method, which is a standard way to solve large-sparse eigenvalue problems. As mentioned previously, our problem is nonlinear eigenvalue problem, so that we have additional density and potential function update processes in the flow chart to achieve the self-consistent solution.

The computational cost of the RSDFT is proportional to the cube of N_{ION} , the number of ions (atoms) in the system. However, the most of computations in $O(N_{ION}^3)$ routines (e.g. Gram-Schmidt) can be performed with level 3 BLAS, so that the computational time is not so much increased as the system size increases.

4 Results of electronic structure calculations for silicon nanowires

SiNW Field-Effect Transistors (FETs) are expected to be boosters in post-scaling semiconductor technology [7]. Clear scaling of short-channel effects vs. NW sizes with the fixed gate lengths has been observed [8]. Reducing the dimensions of SiNWs has been shown to improve the short-channel control. Furthermore, SiNW FETs with channel dimensions of $5.0 \text{ nm} \times 6.3 \text{ nm}$ have been fabricated [8]. A recent experiment showed that the optimal dimension of the cross section of the SiNW FET is around 10 nm [9]. At such small scales, quantum confinement becomes prominent, and therefore reliable quantum mechanical calculations are highly desired to investigate the characteristics of SiNW devices.

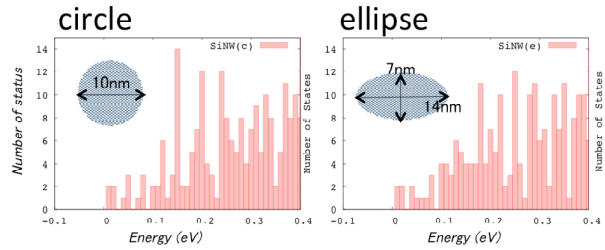


Fig. 3 Dependence of electron states on the morphology of the cross sectional shapes of the silicon nanowires.

We have studied the electron states of SiNWs with several cross-sectional shapes and channel lengths. The size of the systems is ranging from 10,000 atoms to 100,000 atoms. These numbers of atoms are the same as those of real device sizes of SiNWs used for surrounding-gate type FETs.

In Fig. 3, we show the calculated electron density of states of SiNW with circular and elliptic cross sectional shape. Clearly, the electron states depend on the cross-sectional shape of SiNWs due to quantum confinement effect.

From the first-principles calculations, we can calculate the current and current-voltage characteristics of SiNW FETs. These are helpful information for the development of real devices. We believe that the RSDFT code opens a way to study SiNW FETs from first-principles of quantum mechanics in real device sizes.

5 Summary

We have developed RSDFT code to perform large-scale first-principles quantum mechanical calculations on massively parallel computers. RSDFT efficiently works on the K computer, and it makes calculations for 10,000-atom to 100,000-atom systems possible. We got Gordon-Bell prize in 2011 with this work.

We perform first-principles calculations for SiNWs in real-device sizes, and confirm the dependence of electron states on the cross-sectional shape of SiNW due to quantum confinement effect. The results show that RSDFT opens a way to study SiNW FETs from first-principles.

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