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Development of the simulation package 'ELSESES' for extra-large-scale electronic structure calculation

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Abstract

An early-stage version of the simulation package 'ELSESES' (extra-large-scale electronic structure calculation) is developed for simulating the electronic structure and dynamics of large systems, particularly nanometer-scale and ten-nanometer-scale systems (see www.elses.jp). Input and output files are written in the extensible markup language (XML) style for general users. Related pre-/post-simulation tools are also available. A practical workflow and an example are described. A test calculation for the GaAs bulk system is shown, to demonstrate that the present code can handle systems with more than one atom species. Several future aspects are also discussed.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Process (molecular dynamics, MD) calculations with electronic structure are essential as analysis or prediction tools for nanomaterials, particularly materials on nanometer or ten-nanometer scales. Since the structure and function of materials on these scales are determined by the competition among different regions, typically surface and bulk (inner) regions, the theory should reproduce such a nanoscale competition by describing correct electronic structures in different regions. For years, we have developed a set of theories and program codes for such nanoscience research [1–9]. One crucial point is that large-scale quantum-mechanical calculation can be realized, in principle, by calculating the one-body density matrix, instead of one-electron eigenstates, since the computational cost can be drastically reduced [10]. An overview of these theories can be found in the introduction part of [7]. Practical methods were constructed as methods for obtaining the one-body density matrix or the Green's function for a given Hamiltonian matrix. We note that some of the theories are purely mathematical ones, iterative linear-algebraic algorithms for large matrices and, therefore, should be useful in other fields of physics. Actually, one method, called the 'shifted conjugate-orthogonal conjugate-gradient method' [6], was applied to an

extended Hubbard model for $\text{La}_{2-x}\text{Sr}_x\text{Ni}_2\text{O}_4$ [11]. Another crucial point is constructing algorithms for efficient parallel computations. Since multi-core CPU architectures are now built into standard workstations or personal computers, parallel computations are essential for actually all computational systems. The calculations are realized with Slater–Kosterform (tight-binding) Hamiltonians and test calculations were carried out with 10^2 – 10^7 atoms with or without parallelism. See figure 1(a) for a benchmark, in which the parallelism was realized through the OpenMP directive (www.openmp.org). As a benchmark, with a recent multi-core CPU architecture, we have tested our code with a standard workstation with four dual-core CPUs (Opteron 2GHz), for liquid carbon with 1728 atoms. We adopted a typical Hamiltonian of a carbon system [12]. The resulting computational time is six seconds per time step in the process (MD) calculations and the parallel efficiency is more than 90%. Electronic properties, such as the density of states, are also calculated [7].

Now the code has been named the extra large-scale electronic structure calculation (ELSESES) code (www.elses.jp). It is reorganized as a simulation package with input/output files in the extensible markup language (XML) style (<http://www.w3.org/XML/>), as shown in figure 1(b), for a wider range of

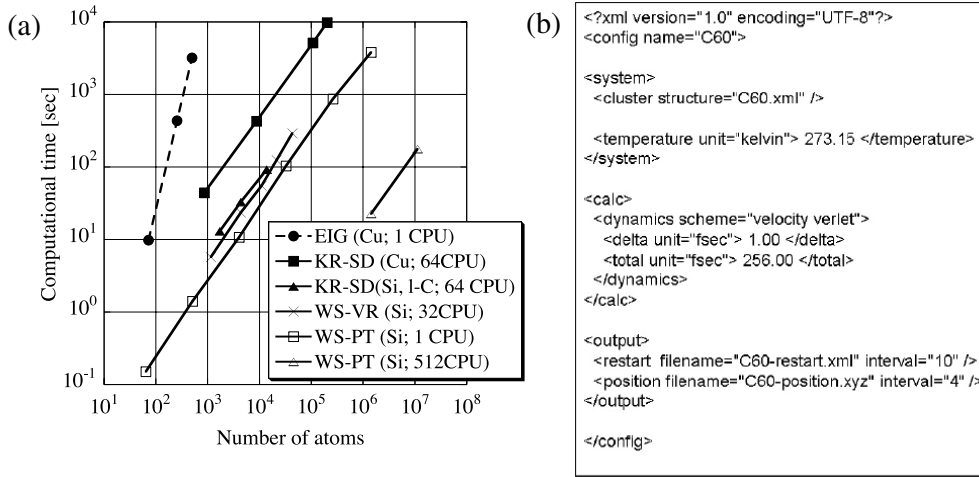


Figure 1. (a) The computational time as a function of the number of atoms (N) [2, 5, 7]. The time was measured for metallic (fcc Cu and liquid C) and insulating (bulk Si) systems with up to 11 315 021 atoms, through conventional eigenstate calculation (EIG) and by our methods for large systems: the Krylov-subspace method with a subspace diagonalization procedure (KR-SD), and the Wannier-state method with variational and perturbative procedures (WS-VR, WS-PT). See the original papers [2, 5, 7] for details of the parallel computation. (b) An example of an XML formatted input file. In this example of an input file, calculation conditions are set as: initial atomic structure file: C60.xml, integration algorithm for molecular dynamic calculation: velocity Verlet, time step: 1.0 fs, total simulated time: 250 fs. Results for atomic structures are written in a file named ‘C60-position.xyz’ every four steps, and the restart file ‘C60-restart.xml’ is updated every ten steps.

users and applications. This article describes the structure, an example and future aspects of the simulation code.

2. Structure of the code

The structure of the code is illustrated schematically in figure 2. The outer loop of the code is the loop of time evolution in the MD simulation (MD part) according to the Newton equation:

$$M_I \ddot{\mathbf{R}}_I = \mathbf{F}_I \quad (1)$$

where M_I , \mathbf{R}_I are the mass and position of the I th atom (ion), respectively, and \mathbf{F}_I is the force on the atom.

The time evolution is performed numerically by a finite-difference method, as in classical MD simulations. At each time step, the quantum-mechanics (QM) part is called from the MD part, in which the force is calculated from the position of atoms:

$$\{\mathbf{R}_I\} \Rightarrow H \Rightarrow \rho \Rightarrow \{\mathbf{F}_I\}. \quad (2)$$

Here H is the Hamiltonian matrix and ρ is the one-body density matrix, defined formally as

$$\rho(\mathbf{r}, \mathbf{r}') \equiv \sum_i f_i \phi_i^*(\mathbf{r}) \phi_i(\mathbf{r}'), \quad (3)$$

from eigenstates $\{\phi_i(\mathbf{r})\}$ and their occupation number $\{f_i\}$. The diagonal elements ($\mathbf{r} = \mathbf{r}'$) give the electron density ($n(\mathbf{r}) \equiv \rho(\mathbf{r}, \mathbf{r})$) and the off-diagonal ones ($\mathbf{r} \neq \mathbf{r}'$) are responsible for the quantum-mechanical effect. In an order- N calculation, such as a Krylov-subspace method [4, 7], the density matrix is calculated without eigenstates $\{\phi_i(\mathbf{r})\}$. In the present article, the method of calculation of the density matrix from the Hamiltonian ($H \Rightarrow \rho$) is called the ‘solver’. In the

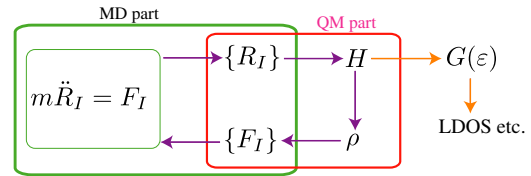


Figure 2. Illustrated structure of the code.

practical code, the matrices are given within the atomic orbital representation:

$$H(\alpha, \beta, I, J) = \langle \alpha, I | \hat{H} | J \beta \rangle \quad (4)$$

$$\rho(\alpha, \beta, I, J) = \langle \alpha, I | \hat{\rho} | J \beta \rangle \quad (5)$$

where the suffices I, J denote atoms and the suffices α, β denote orbitals. The local density of states (LDOS) is calculated through the Green function $G(\epsilon)$, with a Krylov-subspace solver [6]:

$$H \Rightarrow G \Rightarrow \text{LDOS}, \quad (6)$$

which is carried out using a post-simulation tool.

Figure 3 indicates the workflow in research. Two input files in the XML style are needed: (a) file for calculation conditions (see figure 1(b)) and (b) a file for the initial atomic structure. Several conversion tools, such as pre-simulation tools, are available so as to create the XML-style file of atomic structure from non-XML-style files, such as files in the conventional XYZ style.

The XML style is used commonly for text-based electronic files for sharing data through the internet. It is flexible and extensive, because it allows us to define our own items.

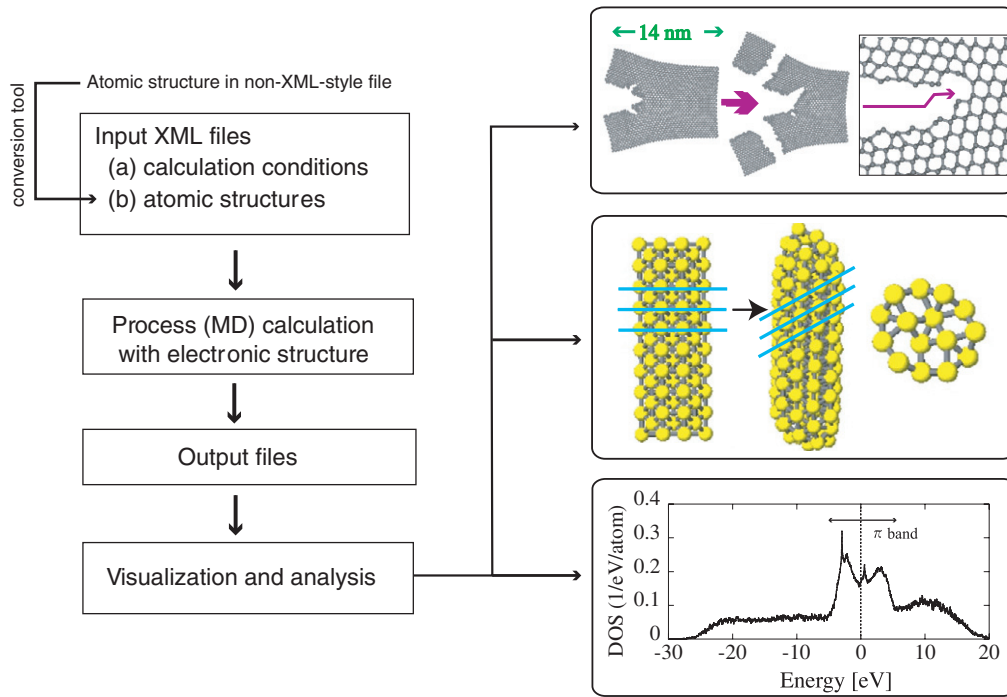


Figure 3. Schematic figure showing the workflow of research with examples of results (right panels). As examples, the silicon cleavage process [5], formation process [8] for helical multi-shell gold nanowire [13], local density of states of liquid carbon [7] appear, in the upper, middle and lower panels on the right, respectively.

In an XML-style file, each item is designated by a start tag ($\langle \dots \rangle$) and an end tag ($\langle / \dots \rangle$). As an example from our XML-style file, figure 1(b) contains a line ' $\langle \text{temperature unit} = \text{'kelvin'} \rangle 273.15 \langle / \text{temperature} \rangle$ ', which means that the temperature of the system is set to be $T = 273.15$ K. Other units can be used by rewriting the 'unit' part, such as 'unit = 'eV'', for eV.

We note that the implementation of an XML-style input file is important in practical simulations of nanostructure materials, since various conditions are required. For example, the fracture simulation of silicon nanocrystal [5] was realized by imposing an external load on the atoms in a limited region near the sample boundary. The extensibility of an XML-style file can satisfy these detailed conditions, by adding newly defined tags for its own purposes.

After the MD simulation, we analyze the atomic processes and electronic structure in detail as well as visualizing atomic structures. The LDOS calculation tool is available as a post-simulation tool, as discussed above. The tools should be developed further, particularly for analyzing electronic structure, such as crystal orbital Hamiltonian populations [14, 6] and a method of quantitative visualization of a chemical bond from the energetics with off-site elements of the Green's function $G(\epsilon)$.

3. Test calculation for a compound; the example of bulk GaAs

A calculation was done for bulk GaAs so as to demonstrate that the present code can handle systems with more than one atom species. We adopt a Slater–Koster-form Hamiltonian for

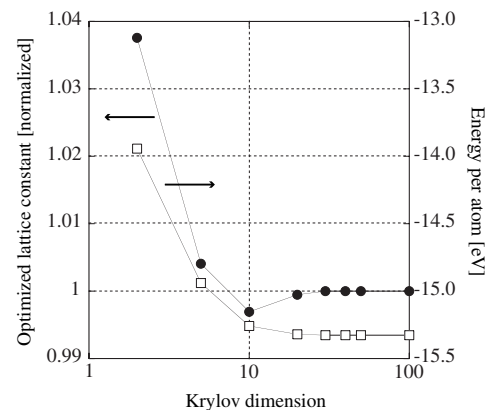


Figure 4. The optimized lattice constant and the energy for bulk GaAs as a function of the Krylov dimension.

GaAs with s, p and s^* atomic orbitals [15]. The atomic energy level of the s^* orbital is located within the conduction band and its physical origin is a spherical average of the five d orbitals. The formulation of s, p, and s^* orbitals was introduced [16], among various semiconductors, for reproducing the valence band and the bottom of the conduction band and was used in papers such as [15, 17–22] for liquid, amorphous, defect, surface and quantum dot cases. Figure 4 shows calculation results for bulk GaAs, in which the cubic periodic cell with 64 atoms is used. Here the Krylov-subspace method with subspace diagonalization [4, 7] is adopted for the solver routine for the density matrix. The dimension of the Krylov subspace (Krylov dimension) should be set as a controlling parameter

that determines the accuracy and computational cost. The computational time is proportional to the Krylov dimension and the calculation will be converged to the exact one when the Krylov dimension increases. See [7] for details. Figure 4 plots the optimized lattice constant and the energy as functions of the Krylov dimension. Figure 4 indicates that the calculation is well converged with the Krylov dimension of 30: the deviations in the lattice constant and the energy are less than 0.01% and less than 1 meV per atom, respectively. We note that an excellent convergence at the Krylov dimension of 30 was found in the other systems [4, 7].

4. Summary

A large-scale electronic structure calculation code is being developed as a simulation package with the name of ELSESES (www.elses.jp). For a better user interface of our simulation code, we have created input/output interfaces of XML-style files. The pre- and post-processing tools have been also prepared for modeling and detailed analysis of the atomic structures. We have also confirmed that the present code can handle system more than one atom species by making a calculation for bulk GaAs. Although the present stage of the simulation package is still early, we believe that the code will provide fruitful simulations for researchers in nanomaterial science. As a future aspect, the method is being extended by implementation of general Slater–Koster-form Hamiltonians for a wider range of materials, in which an explicit charge self-consistent treatment [23] is included. Non-equilibrium current and other electronic properties are also crucial for nanoscience and should be investigated with the present methodologies.

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