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An efficient hybrid molecular dynamics simulation method for solids*

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An efficient molecular dynamics simulation method for solids is presented, where two different schemes are hybridized. This hybrid method is implemented in spatially distributed computer systems, and can be processed at high speed over 10 times in comparison with conventional simulation methods.
Key words: hybrid algorithm, molecular dynamics, ONIOM

1. INTRODUCTION

A molecular dynamics (MD) simulation is a powerful tool for investigation of a mechanical property of solids. In the MD simulation, constituent atoms are moving according to forces acting on each atom, and the forces are evaluated from inter-atomic potentials. If the inter-atomic potentials are assumed as simple potential forms derived from a tight-binding Hamiltonian, the MD simulation can be carried out very easy, and, therefore, a very large system containing $\sim 10^6$ atoms can be analyzed by the MD simulation. However, the tight-binding Hamiltonian contains several adjustable parameters, and there is an ambiguity to determine these parameters. Recently, MD simulations based on a fully quantum mechanics have been developed, and these MD simulations have no adjustable parameter, which called *ab-initio* molecular dynamics (AIMD) simulation [1]. Since the AIMD simulation has no empirical parameter, the AIMD predicts electronic, mechanical and other properties of materials without any experimental data. But, unfortunately, there is a limitation for application of the AIMD, because the AIMD simulation exhausts a large computational time.

The AIMD simulation gives a highly reliable result without empirical parameters, and requires a huge computational times. On the other hand, the tight-binding molecular dynamics (TBMD) simulation using simple inter-atomic potential is able to deal with a large system containing a lot of atoms and molecules, in which several empirical parameters are to be determined. It is expected to perform an efficient simulation by using the AIMD together with the TBMD simulations, which called a hybrid MD simulation. In the hybrid MD, the AIMD is used in a small region containing chemically reactive part, while the TBMD is carried out in a large region where there is no chemically reactive part. It is necessary to calculate the forces acting on each atom based on quantum mechanics in the chemically reactive region, and, on the other hand, it is enough to evaluate the forces acting on each atom via parameterized inter-atomic potentials in the chemically stable region.

Morokuma and his coworkers have pre*sented an efficient hybrid scheme to evaluate energy of a large isolated molecule and its derivatives, i.e. the forces acting on atoms, which called ONIOM method [2,3,4]. Morokuma group has shown good numerical results for many large molecules including biomaterials. The present author expands the ONIOM method into a hybrid MD simulation scheme for solids. In our scheme, two different approximations to construct Hamiltonian, i.e. density functional theory (DFT) and parameterized tight-binding theory, and two different sized atomic structures are incorporated under a periodic boundary condition. The numerical test of this method is presented in this paper.

2. METHODOLOGY

The present method is based on a supercell method. In conventional supercell method, a large-sized supercell is needed to obtain a precious numerical result. In order to avoid handling such a large-sized supercell, another small-sized supercall is introduced. This small-sized supercell corresponds to a small region in the large-sized supercell, which contains chemically reactive region like a defect, impurity, reactive site, and so on. According to the original ONIOM method, the large-sized and small-sized supercells are named as *REAL* and *MODEL* systems, respectively. We denote two different approximations to construct the Hamiltonian as *HIGH* and *LOW*, which means the DFT and parameterized tight-binding methods, respectively. Thus, in similar way to the ONIOM method [2], the total energy $E(HIGH,REAL)$ is approximately given by

$$E(HIGH,REAL) \approx E(LOW,REAL) - E(LOW,MODEL) + E(HIGH,MODEL). \quad (1)$$

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Differentiating the E by atomic coordinates $\boldsymbol{Ra}$, we obtain the force acting on the atom a,

$$\boldsymbol{Fa} = \frac{\partial E}{\partial \boldsymbol{Ra}} \approx \frac{\partial E(LOW, REAL)}{\partial \boldsymbol{Ra}} - \frac{\partial E(LOW, MODEL)}{\partial \boldsymbol{Ra}} + \frac{\partial E(HIGH, MODEL)}{\partial \boldsymbol{Ra}} \quad (2)$$

In Equations (1) and (2), the computational time of the third term in the right hand is dominant in a whole computational time. In original ONIOM formulation [2], link atoms are introduced to terminate the surface atoms of the *MODEL* system. However, in the present formulation for solids, the link atoms are unnecessary, so that Jacobian matrix does not appear in Equation (2).

The present hybrid MD becomes a coarse grain algorithm, so that this scheme is adequate to be executed on spatially distributed complex computers. Because the AIMD consumes huge computational times, it is desirable that the AIMD is carried out on a supercomputer, but the other part can be evaluated by using PC's. In this implementation, amount of the communication data between computers is small, it is enough to employ usual 100-based LAN system or INTERNET.

3. NUMERICAL TEST

The numerical result for a defect energy in Si is presented in Table 1, where the electronic structures of *MODEL* systems are calculated in the framework of DFT with the local approximation for the exchange-correlation energy, and the electronic structures of *REAL* systems are calculated by using parameterized tight-binding Hamiltonian. The former calculations are pursued by using plane wave basis with the cut-off energy of 8Ryd. The later calculations are carried out non-selfconsistently, and, thus, are performed quickly. If the *MODEL* system containing 64 atoms and the *REAL* system containing 1000 atoms are adopted as the hybrid system, calculated defect energy is 3.9023eV. The error from the energy obtained by using the large-sized superell consisting of 216 atoms is about 1%, and CPU time required for the hybrid system is about 1/50 in comparison with the large-sized supercell. The present hybrid MD simulation method is highly efficient for semiconductors. Now we are checking the efficiency of the method for metallic systems, which will be reported in elsewhere.

Table 1. Point defect energy in silicon . Error means difference from the *ab-initio* defect energy estimated by using the large supercell containing 216 atoms.

Number of atoms in *MODEL* system	Number of atoms in *REAL* system	Defect energy (eV)	Error (eV)
8	64	2.5684	1.3745
8	216	3.1525	0.7904
8	512	3.2454	0.6975
8	1000	3.2712	0.6717
64	216	3.7836	0.1593
64	512	3.8765	0.0664
64	1000	3.9023	0.0406

4. CONCLUDING REMARKS

A hybrid MD simulation method for solids based on the ONIOM scheme for large isolated molecules has been presented, and has given an efficient and good result for evaluation of defect energy in Si with lattice relaxations. This hybrid MD simulation method is coarse-grained from the viewpoint of parallel and distributing processing, and, therefore, is suitable for distributed computing, even if the communication speed between processor elements is slow. Therefore the hybrid MD simulation method is applicable for a large area computer network and GRID computing.

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