Large Scale Simulations for Carbon Nanotubes

Project Representative

Kazuo Minami Research Organization for Information Science &Technology

Authors

Syogo Tejima *1, Kazuo Minami *1, Yoshiyuki Miyamoto *2, Mari Ohfuti *3
Mamoru Sakaue *3, Tsuyoshi Miyazaki *4 and Hisashi Nakamura *1

*1 Research Organization for Information Science &Technology (RIST)

*2 Fundamental and Environmental Research Laboratories, NEC Corporation

*3 Nanotechnology Research Center, Fujitsu Laboratories Ltd.

*4 National Institute for Materials Science (NIMS)

CARBON NANOTUBE RESARCH GROUPE

Morinobu Endo, Eiji Osawa, Atsushi Oshiyama, Yasumasa Kanada, Susumu Saito,
Riichiro Saito, Hisanori Shinohara, David Tomanek, Tsuneo Hirano,
Shigeo Maruyama, Kazuyuki Watanabe, Ohno Takahisa, Yoshiyuki Miyamoto
and Hisashi Nakamura

Carbon nanotubes (CNTs) and fullerenes have a lot of potential applications in nanotechnology. In the stream of efforts to exploit these nanoscale materials, the computational simulations have turned out to be powerful and efficient tools. Especially, as the recent trend in technology made it possible to manipulate further miniaturized structures, required simulations for emerging material become bigger and bigger. Aiming at realistic simulations for nanomaterials, we have developed a large-scale computation technique utilizing tight-binding molecular dynamic method, *ab initio* density functional theory (DFT), and time-dependent DFT method. By an efficient optimization on ES, we achieved the performance of 7.04 Tera flops using 435 processors.

We have studied various physical properties of nano-carbon material e.g., hot carrier dynamics, atomic and electronic structure of CNT-Metal contacts, structure of bundle CNTs, and creation of novel nanocarbon structures. During these works, we have realized that the Earth Simulator is a very powerful tool for large-scale material simulations.

Keywords: Large-scale simulation, TB theory, *ab initio* theory, DFT, Carbon Nanotube, Fullerens

1. INTRODUCTION

Carbon materials have been expected to make a breakthrough in material science and nanotechnology. A lot of potential applications of nanotubes and fullerenes e.g., electronic field emitter and electronic devices have attracted a scientific community. In the investigation and utilization of their material properties, numerical simulation using a supercomputer has turned to be a very efficient tool. A recent development in nanotechnology has required more efficient supercomputing capable of large-scale simulation of up to 10^4 atoms.

Aiming at large-scale simulation utilizing the Earth Simulator, we have developed computational software packages based on *ab initio* DFT theory and parameterized tight-binding (TB) method. The TB code we have developed is shown to be suitable for very large systems in spite of the lack of symmetrical arrangement. We have carried out some subjects in this work, which are described in the next section. There are three primary objectives: design of innovative nanomaterials with certain desired properties; obtaining fundamental properties in nano-scale matter, and nano-applications. Our purpose is to give the clear explanation of properties and phenomena of nano-scale events and deduce guiding principle to design new materials from nano-structures using super-computers.

2. PHYSICAL STUDIES ON NANOMATERIALS

2.1. Hot carrier dynamics in carbon nanotube

In 2003, we started the simulation on hot-carrier decay in carbon nanotubes which enables us to extract time-constant of carrier decay and this parameter will be crucial for designing optimum running frequency of carbon nanotube devices, such as transistor, and optical switch. We obtained rapid decay of the energy gap of excited electron-hole pair

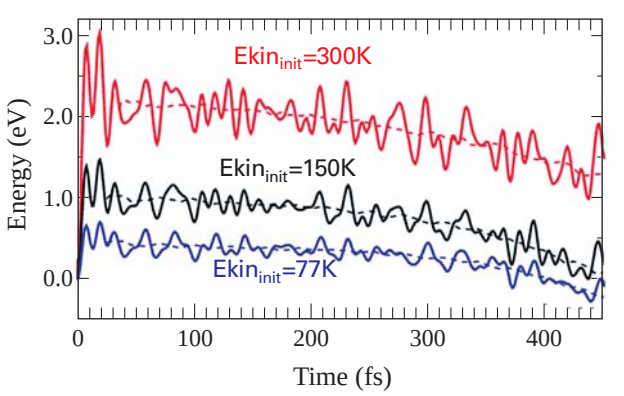

Fig. 1 Time-evolution of the potential energy of ionic system in electronically excited carbon nanotubes. Lower initial temperature gives slower drift of the potential.

which can be interpreted as early electron-electron decay and later electron-phonon coupling.

In this year, we have confirmed temperature-dependence of the electron-phonon coupling appeared in the later time-domain. The temperature dependence is mimicked by giving a certain scale of the randomized velocities on all ions in the beginning of the simulation in accordance with the Maxwell-Boltzmann distribution function. Then we compare the time-evolutions of the potential energy of ionic system with different scales of the initial velocities on ions. Figure 1 shows temperature dependence of the potential lowering. One can note that lower temperature case shows slower drift of the potential which means slower rate of energy transfer from the electronic system to ionic system. The dotted lines are time-average of the fluctuating potentials which highlight the trend of the potential drift. Corresponding paper is now under preparation.

2.2. Atomic and Electronic Structures of CNT-Metal Contacts

We have studied the atomic and electronic structures of CNT-metal contacts by using *ab-initio* molecular dynamics.

While the systems are stabilized from the simple contacts, both the structures of CNT-molybdenum contacts remarkably change, compared with CNT-titanium contacts. Although CNTs have shown potential properties for electronic devices, the contacts with the metal, which would play important roles in the applications, have not been understood enough. The atomic and electronic structures of CNT-metal contacts have been studied by using *ab-initio* molecular dynamics. We have examined titanium (Ti) and molybdenum (Mo) as possible contact materials, because they are usually used in the layers under catalysts for CNT growth. The initial atomic structure is formed by simply connecting metallic (5,5) CNTs with metal slabs. The unit cell has 90 carbon atoms and 96 metal atoms. Both Ti and Mo in the simple contacts give good electronic structures, where the electronic states of the metal distribute around the Fermi level of CNT. When the system is stabilized, the atomic structure of the CNT-Mo contact remarkably changes, compared with the CNT-Ti contact. The remarkable change in the atomic structure also brings a change in the electronic structure. The density of states around the Fermi level is obviously increased, which seems to suggest that Mo is better as a contact material. We also will investigate semiconductor and multi-walled CNTs.

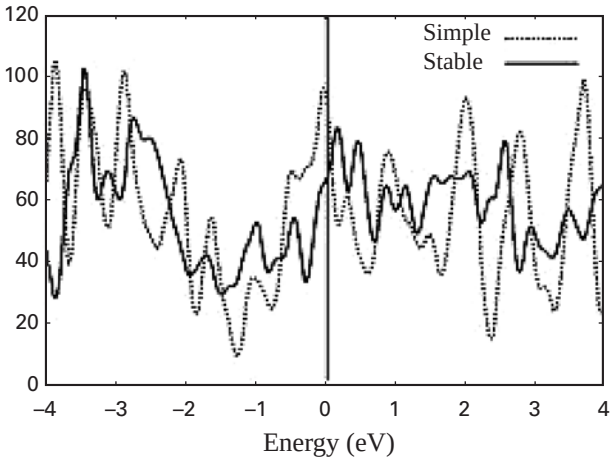

Fig. 3 Density of states of CNT-Ti contacts.

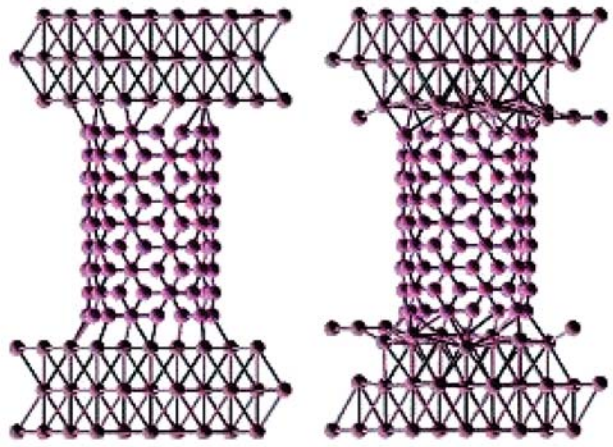

Fig. 2 Simple and stable atomic structures of CNT-Ti contacts.

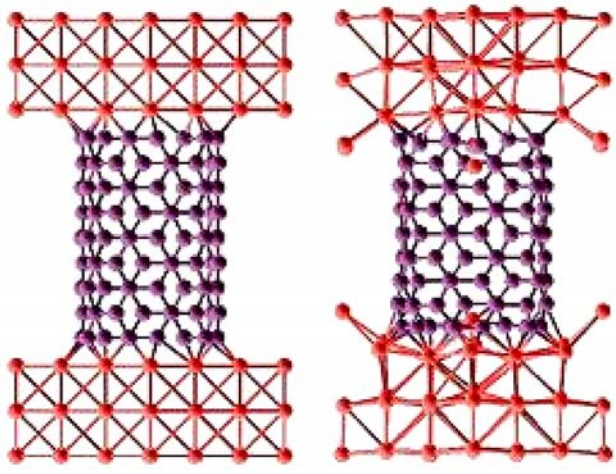

Fig. 4 Simple and stable atomic structures of CNT-Mo contacts.

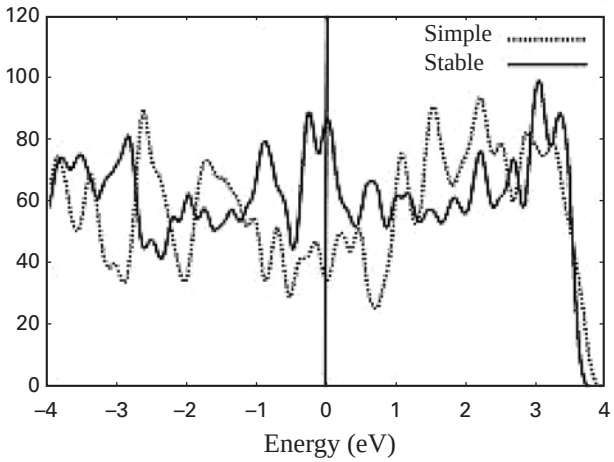

Fig. 5 Density of states of CNT-Mo contacts.

2.3. Advantaged order-N first-principle method for large complex systems.

Conventional first-principles calculations based on the density functional theory (DFT) are very inefficient for large complex systems because the memory and CPU requirements increase rapidly as the number of atoms in the target systems increases. In order to overcome this problem, we are developing a linear-scaling DFT code, which enables us to perform DFT calculations on very large systems. Within this fiscal year, we have optimized the code for the Earth Simulator and have demonstrated its high performance especially in parallelization. We have succeeded in performing DFT calculations on the system of three-dimensional Ge overlayers on Si (001), which includes about ten thousands of atoms. The present parallelization ratio is 99.91% and we can expect even higher values when we treat larger systems.

2.4. Structure of bundle CNTs for composite materials

In 2003, we reported mechanical properties as pull and push of single- and double-CNT using the large scale tigth-binding simulation. In that works, we presented the structure dependence on collapse and fracture in CNTs.

When we used the strong nanotube as a real material, it was necessary to bundle these CNTs. As a first stage, the effect of Van der Waals attractive interaction between the different number of CNTs to bundle together was simulated under no applied forces. Two types of bundle CNTs are placed at triangle lattices at an appropriate distance between CNTs (Fig. 6, 7). When each CNTs tried to approach together by the attractive force, then faces of contact become flat (Fig. 6 (1, 2, 3) and Fig. 7 (1, 2, 3)).

In spite of same simulation time steps, the bundle consisting of a small number of nanotubes shown in Fig. 6 looks like structure separation as a reaction. We predicted that binding of CNTs could be stronger with increasing CNTs.

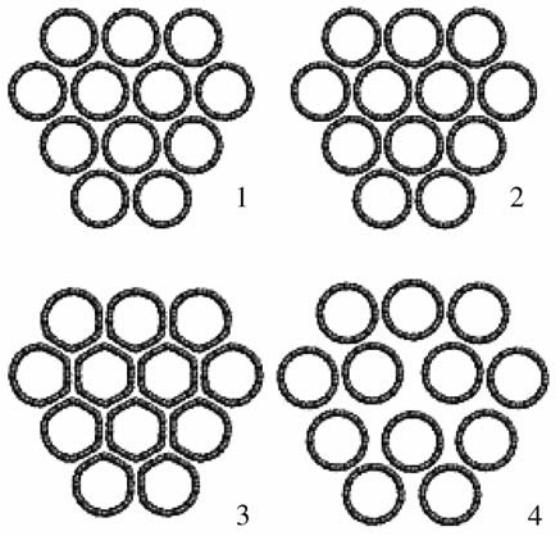

Fig. 6 Creation process of bundle by twelve nanotubes

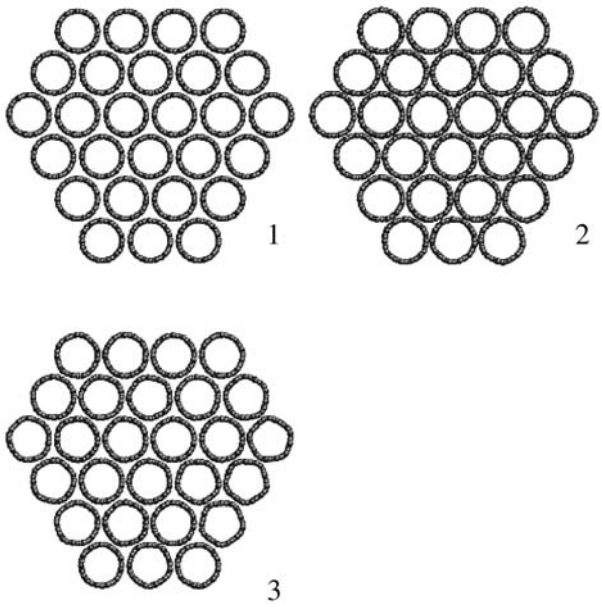

Fig. 7 Creation process of bundle by twenty-seven nanotubes.

2.5. Creation of novel nanocarbon structures using GSW rearrangement

This work is on quantum combinational chemistry simulation. Its objective is to find new nanocarbon structure by isomerization using generalized Stone-Wales (GSW) rearrangement and C2 loss. Isomerization is extremely compute intensive, so the simulation clearly needs very large-scale computers. There is a parallel code using a master-slave algolism. One processor as master is gather and classify all reaction paths generated on other processors as slave. The fig.8 shows the generation of a capped long CNT from a short CNT and a fullerene. The Fig. 9 exhibits the nanotube generation from three fullerenes applying the boundary periodic condition. The zigzag and one chilral nanotube appear in the simulations. The long-term goal is to find a path to make new nanodevices from the many generated nano structures by self-organization.

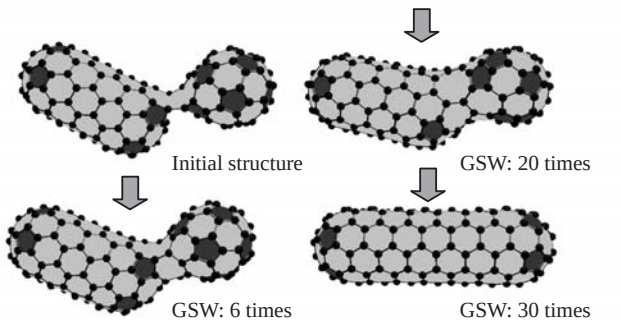

Fig. 8 Generation of capped CNT from two fullerenes.

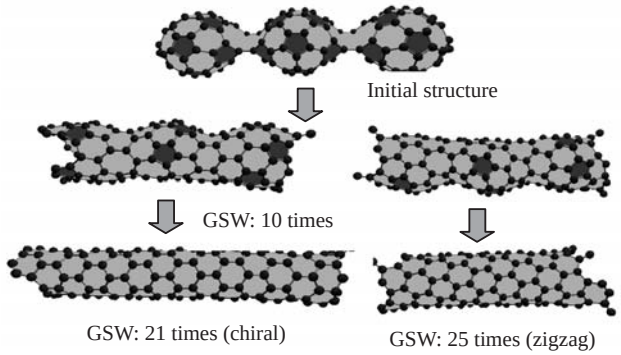

Fig. 9 Generation of CNTs.

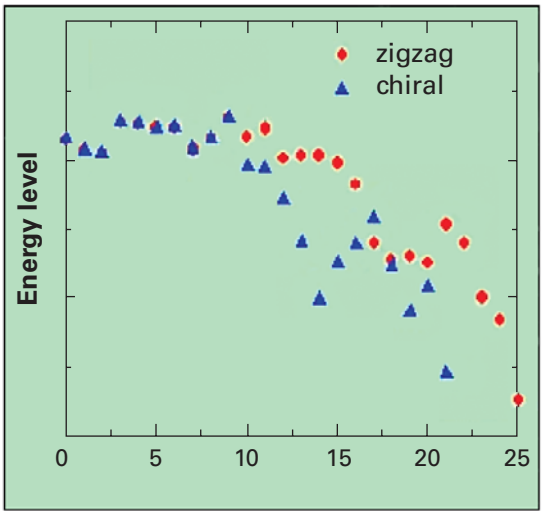

Fig.10 Energy v.s number of GSW sequence

3. SUMMARY

The large-scale simulations on nanomaterials have been carried by *ab initio* density functional method and the parameterized out tight-binding calculations. The optimized codes showed that the computation on the Earth Simulator could give an exceptional performance and enable more large-scale realistic simulations. Our large scale simulations for nano applications will contribute to valuable on novel nano material properties and on nano electrical designs.

カーボンナノチューブの特性に関する大規模シミュレーション

プロジェクト責任者
南　　一生　　財団法人 高度情報科学技術研究機構

著者
手島　正吾*1，南　　一生*1，宮本　良之*2，大淵　真理*3，坂上　　護*3，宮崎　　剛*4，
中村　　壽*1

*1　財団法人 高度情報科学技術研究機構
*2　NECラボラトリーズ 基礎研究所CNT応用研究センター
*3　株式会社富士通研究所
*4　物質・材料研究機構 計算材料科学研究センター

概要

1. 研究目的

優れた物性と予想されナノテクの基本材とされるカーボンナノチューブ(CNT)類の電子・機械特性を従来不可能だった大規模シミュレーションにより推定し、科学技術及び産業界に提供すると共に、応用として特性の優れた新構造を発見し、基本材の拡充に寄与する目的で実施された。

2. 成果

H14年度、地球シミュレータを利用した大規模シミュレーションの有効性を世界に先駆けて実証した。またH15年度は、我が国のナノチューブ研究で当面する課題解決に向けた応用シミュレーションを実施した。本年度さらに、応用シミュレーションを推進し得られた成果を以下に示す。(1)基本特性把握の面から、産業界で必要となるナノチューブのバンドル構造のシミュレーションを開始した。本シミュレーションは量子計算では世界最初となり、バンドルの大きさが増すと凝縮の度合いが強まっていく傾向が把握された。(2)次世代回路応用特性把握の面からナノチューブの光デバイス動作速度のシミュレーションおよびその温度依存性シミュレーションを実施し、動作速度は温度に敏感である事を明らかにした。またナノチューブ・金属結合の電子特性シミュレーションを実施し、ナノチューブと金属の接合における原子構造の安定性をシミュレーションした。(3)さらに新物質創製のためのナノ構造加工特性把握の面から、C60からナノチューブができる可能性を探るGWS転移シミュレーションを実施した。

キーワード：大規模シミュレーション，タイトバインディング理論，アビニシオ理論，密度汎関数法，カーボンナノチューブ，ナノダイヤモンド