博　士　論　文　（　要　約　）

Development of O(N) *Ab-initio* Molecular Dynamics that Exploits the Locality of Forces and its Acceleration through Machine Learning

（力の局所性を利用したオーダーＮ・第一原理分子動力学法の開発及び機械学習によるその高速化）

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In the conventional research conducted by *ab-initio* calculations, the atomic structure of the target system is often unknown *a priori* and huge amount of the calculation time is spent on the determination of the stable atomic structures before the truly interesting investigation of the electronic structures. As the calculation cost of the conventional density functional theory (DFT) grows as a cube of the target system size N (order N^3; $O(N^3)$), the scaling order of the calculation is the bottleneck to extend its application to larger realistic systems. Thus, alternative methodologies to obtain the stable structures are desirable. Furthermore, due to the increase in the number of (meta-)stable structures as the target system grows, simple geometry optimization techniques in the standard DFT tend to be stuck to these local minima. Techniques such as simulated annealing work well even with those kinds of situations.

Here I propose a new way of the generation of molecular dynamics (MD) simulation with O(N) scaling by exploiting the locality of atomic forces to trade computational accuracy with computational cost. The propagation of the effect of the atomic forces is empirically known to be limited spatially, leading to the success of various classical force fields. In the new method, the atomic forces are calculated from auxiliary subsystems, and the obtained forces are used to perform the MD simulation. The error in the forces induced by the auxiliary subsystems can be systematically reduced by increasing the size of the subsystems.

First, the locality of the atomic force is analyzed with the assumption that the contribution to the forces of a given atom from the surrounding atoms is independent. This independence *ansatz* leads to the locality of the atomic force analytically and it is

confirmed numerically with the calculation on various systems including, bulk silicon, bulk silicon carbide, bulk silicon dioxide, and bulk aluminum. Furthermore, I have shown that with an appropriate hydrogen-termination treatment of the subsystem surface and some randomness in the subsystem selection, the forces are obtained with good agreement to the conventional $O(N^3)$ DFT and also a good conservation in the total energy during the MD simulation is achieved. This can be intuitively explained by that with the randomness, the systematic error induced by the subsystem in the atomic forces is reduced throughout the time evolution. The calculated phonon DOS of silicon from the molecular dynamics trajectory by the proposed method shows good agreement with the ones by the conventional *ab-initio* methods. Unlike electron density or locally defined energy, the atomic forces are assigned to each atoms without ambiguity. This feature leads to one advantage of the proposed method that the method is mobile in the sense that it can be easily used with any other force fields. This enables an easy extension of QM/MM scheme in phase space where the quantum region and the molecular region can be mixed in both spatial and temporal regimes. The classical force fields are considered to replace some parts of the *ab-initio* calculations to further reduce the computational cost.

For our original motivation of the exploration of stable structures with the simulated annealing, the geometry optimization of SiC surface oxygen adsorption is performed. The energy difference between the final structures can be well reproduced with the proposed new method. Also, the systematic improvement of computational accuracy with increasing the subsystem size is shown. The proposed method works well with complex systems containing multiple kinds of elements and surfaces. The typical target system size for the crossover of computational time starts from several hundred atoms. The simulated annealing for the graphene/SiC interface system with around nine hundred atoms is performed.

In summary, an efficient O(N) methods to conduct *ab-initio* MD is developed. It exploits the locality of atomic forces to achieve ideal O(N) scaling. The proposed method can systematically trade computational accuracy with computational cost. The simple division of the problem to obtain atomic forces allow simple hybridization of methods of various classical force fields to further reduce computational cost.