Self-learning Hybrid Monte Carlo: A First-principles Approach

Yuki Nagai,1,2 Masahiro Okumura,1 Keita Kobayashi,3 and Motoyuki Shiga1

1 CCSE, Japan Atomic Energy Agency, 178-4-4, Wakashiba, Kashiwa, Chiba, 277-0871, Japan
2 Mathematical Science Team, RIKEN Center for Advanced Intelligence Project (AIP), 1-4-1 Nihonbashi, Chuo-ku, Tokyo 103-0027, Japan
3 Research Organization for Information Science and Technology (RIST), 2-4, Shirakata, Tokai-mura, Ibaraki 319-1106, Japan

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We propose a novel approach called Self-Learning Hybrid Monte Carlo (SLHMC) which is a general method to make use of machine learning potentials to accelerate the statistical sampling of first-principles density-functional-theory (DFT) simulations. The trajectories are generated on an approximate machine learning (ML) potential energy surface. The trajectories are then accepted or rejected by the Metropolis algorithm based on DFT energies. In this way the statistical ensemble is sampled exactly at the DFT level for a given thermodynamic condition. Meanwhile the ML potential is improved on the fly by training to enhance the sampling, whereby the training data set, which is sampled from the exact ensemble, is created automatically. Using the examples of α-quartz crystal SiO2 and phonon-mediated unconventional superconductor YNi2B2C systems, we show that SLHMC with artificial neural networks (ANN) is capable of very efficient sampling, while at the same time enabling the optimization of the ANN potential to within meV/atom accuracy. The ANN potential thus obtained is transferable to ANN molecular dynamics simulations to explore dynamics as well as thermodynamics. This makes the SLHMC approach widely applicable for studies on materials in physics and chemistry.

Introduction. First-principles molecular dynamics based on density functional theory (DFT-MD) is a powerful tool to simulate a variety of materials in physics and chemistry [1]. However, reducing the computational effort required for DFT-MD remains a key issue for its broader application to phenomena on large length- and time-scales. The use of artificial neural networks (ANN), which imitate DFT energies by machine learning, is seen as a promising solution to this issue [2–5]. The branch of the research about machine learning molecular simulations has grown rapidly in the last decade after an influential paper by Behler and Parrinello [5] laid down a general framework of setting up and training ANN potentials from DFT data sets, and running ANN-MD simulations for condensed matter systems.

The training of machine learning (ML) potentials must be based on sufficient amounts of DFT-derived results to cover all the configuration space, which corresponds to a statistical ensemble in the case of systems in thermal equilibrium. Usually the training sets for ML potentials are chosen before starting ML-MD simulations. Many useful methods, such as generic algorithms [16] and CUR decompositions [17] etc., were suggested for properly choosing training sets. If sufficient care is not taken, it is possible that ML-MD simulations may break down suddenly when the trajectory finds its way into an uncovered part of the phase space, see Fig. 1. Therefore it would be beneficial to establish a way to somehow cover the ensemble space in an automatic manner.

Here we propose a novel approach for training ML potentials based on the exact statistical ensemble at a given thermodynamic condition, e.g., isothermal, isobaric ensembles, using the hybrid Monte Carlo technique [19–21]. This allows one to access exact results out of approximate ML potentials, and, at the same time, create an objective and unique data set for ML training. Importantly, we want to circumvent the use of costly DFT-MD computations as much as possible, which can be achieved by training the ML potential on systems with small sizes and short time scales. Our idea is to use the dual-level HMC method [22,23]. The ensemble is sampled by generating

![Fig. 1. Schematic figure of trajectories in phase space. (a) Trajectories of DFT-MD, (b) ML-MD, and (c) SLHMC. If the ML potential is not well-trained, the ML-MD trajectory might fall outside the range of the DFT ensemble (depicted by a dotted oval), while the accepted SLHMC trajectory always stays inside.](image-url)
trial moves of the trajectory from an approximate ML potential energy surface, which are then either accepted or rejected by performing exact DFT energy calculations at long time step intervals. Note that the ensemble created is theoretically exact irrespective of the quality of the approximate ML potentials. Its efficiency is strongly dependent, however, on the quality of the ML potential, as this affects acceptance ratio. It will be shown herein that the acceptance ratio tends to improve as the ML potential is trained and updated iteratively, which is done automatically on the fly during the computations. Note that the ML potential is used here in an auxiliary manner to produce exact ensembles, which distinguishes our concept from previous works where the ML potential was used as an approximation to imitate exact DFT potentials.

We call our method the self-learning hybrid Monte Carlo (SLHMC) method, since it is akin to the self-learning Monte Carlo (SLMC) method [22]. The SLMC method was recently introduced in the field of many-electron systems to speed up MC simulations by using efficient global updates informed by machine learning techniques [23,24]. SLHMC extends the idea of SLMC to equation-of-motion based moves to enable efficient global updates for atomistic and molecular simulations.

This letter is organized as follows: We first explain the basic theory and computational procedure of SLHMC. We use example calculations on SiO$_2$ (α-quartz) to demonstrate the accuracy and efficiency of the method. Calculations for the phonon-mediated superconductor YNi$_2$B$_2$C are then used to demonstrate the ability to construct accurate ML potentials such that the ML-MD simulations are stable at long times.

**Self-learning Hybrid Monte Carlo Method.** A trial move uses Hamilton’s equations of motion derived from the ML potential energy surface, $V_{ML}$,

$$\dot{p}_i = -\frac{\partial V_{ML}(\{r\}, t)}{\partial r_i}, \quad \dot{r}_i = \frac{p_i}{m_i}, \quad (1)$$

where $p_i$, $r_i$, and $m_i$ are the momentum, coordinates, and mass of the $i$-th atom, respectively. Starting with a random initial momentum generated from the Maxwell-Boltzmann distribution, the equations of motion are solved for a discrete time step, $\Delta t_{ML}$, using a time-reversible and area-preserving algorithm (in the present study, the velocity-Verlet algorithm). The ML potential surface $V_{ML}(\{r\}, t)$ depends on time $t$ as it is trained on the fly, but it is kept constant within the time interval $\Delta t_T$, i.e.,

$$V_{ML}(\{r\}, t) = V_{ML}^n(\{r\}), \quad n\Delta t_T < t < (n+1)\Delta t_T, \quad (2)$$

where $V_{ML}^n$ is the ML potential for the $n$-th update. $V_{ML}$ is trained every $\Delta t_T = n_{DFT}\Delta t_{DFT}$, where $\Delta t_{DFT} \equiv n_{ML}\Delta t_{ML}$ is the interval of acceptance/rejection in the Metropolis algorithm. Thus $n_{DFT}$ is the number of times the DFT energy is computed for training, and $n_{ML}$ is the number of steps in a trial move. The acceptance probability for a trial move within the phase space from $\{p, r\}$ to $\{p', r'\}$ is given by

$$P_{acc}(\{p, r\} \rightarrow \{p', r'\}) = \min\left(1, e^{-\beta(V_{DFT}(\{p', r'\}) - V_{DFT}(\{p, r\}))}\right), \quad (3)$$

where $\beta = 1/T$ is the inverse of temperature and

$$H_{DFT} = \sum_{i=1}^{N} \frac{|p_i|^2}{2m_i} + V_{DFT}(\{r\}). \quad (4)$$

is the Hamiltonian based on the DFT potential energy, $V_{DFT}(\{r\})$. We note that the detailed balance condition is preserved exactly on the basis of the DFT (not ML) potential as long as the ML potential $V_{ML}(\{r\}, t)$ does not change during a trial move.

According to Equations (1) and (3), ML force calculations are required $n_{ML}$ times while DFT energy calculations are required once. SLHMC is computationally efficient when the former is less expensive than the latter, which is usually the case unless we set a huge value for $n_{ML}$. In such a case, $\Delta t_{ML}$ could be chosen to be sufficiently small so as to conserve the ML energy within a trial move. (This situation is different from conventional HMC where the step size should be large to break energy conservation.) When the ML energy is conserved within a trial move, the acceptance probability (3) becomes

$$P_{acc}(\{p, r\} \rightarrow \{p', r'\}) \sim \min\left(1, e^{-\beta\Delta V}\right), \quad (5)$$

where $\Delta V \equiv \Delta V(\{r'\}) - \Delta V(\{r\})$ and $\Delta V(\{r\}) \equiv V_{DFT}(\{r\}) - V_{ML}(\{r\}, t)$. Therefore the accuracy of the ML potential influences the acceptance ratio, and thus the efficiency of the SLHMC method. In practice, it is important that $n_{ML}$ is chosen carefully to give a decent acceptance ratio (more than circa 30%).

In this paper, we use the Behler-Parrinello ANN potentials as ML potentials. We follow the standard ANN training protocol to minimize the mean-square error ($\Delta V(\{r\})^2$). The ANN variables are restarted from the last update $(n-1)$ and optimized using all the DFT energy data up to the current update $(n)$. Training is possible using either only the accepted structures, or both the accepted and rejected structures (the latter is employed here). Since the size of the training data set needed is small ($\sim 2000$ here), the computational cost of training the ANN potential is usually not dominant in SLHMC. As the number of training data increase with increasing $n$, one can skip the training steps after the ANN variables are optimized sufficiently by judging from the average acceptance ratio calculated in SLHMC.

**Demonstration I: Thermodynamics of SiO$_2$.** The SLHMC method was implemented in the PIMD code [33] which has the access to ANN potentials and DFT
The first test was on the thermodynamics of the α-quartz phase of SiO\textsubscript{2} crystal \cite{36}. The SLHMC simulations were at 300 K in the canonical ensemble for a periodic system of 24 Si atoms and 48 O atoms. The DFT calculations used the Perdew-Burke-Ernzerhof (PBE) functional \cite{37}. The projected augmented wave (PAW) method \cite{38} was employed, while the cut-off energy was 500 eV and the sampling points were gamma-point only. The ANN potentials were trained using the limited-memory Broyden-Fletcher-Goldfarb-Shanno (L-BFGS) method \cite{39,40}. We adopt the Chebyshev basis set as a descriptor for atomic environments \cite{11}, and the corresponding parameters are given in Ref. \cite{41}. The step size of ML, DFT and training were chosen to be $\Delta t_{\text{ML}} = 0.25$ fs, $\Delta t_{\text{DFT}} = 5$ fs, and $\Delta t_{\text{T}} = 500$ fs, respectively. The initial guess of the ANN potential was prepared and trained using a short DFT-MD trajectory of 300 steps starting from the crystal structure. This could be done in other ways if the SLHMC run gets stuck at the initial step.

Figure 2 shows that the difference between the ANN and DFT potentials quickly diminishes to about 1 meV/atom on the average as the SLHMC simulation proceeds and the ANN potential is trained on the fly. This accuracy can be ascribed to the fact that the ANN potential has been trained in a confined configuration space corresponding to the exact ensemble at the DFT level. Figure 3 shows that the radial distribution functions (RDFs) obtained from the SLHMC and DFT-MD simulations are identical, as they should be. The RDFs in the SLHMC converge faster than those in the DFT-MD. Note that this is the case even when the ANN potentials are changed during the SLHMC simulation. This demonstrates that the SLHMC method is able to gain statistics on thermodynamic properties while training the ANN potential at the same time.

Figure 4 shows the results of the mean squared displacement (MSD) as a function of the number of DFT calculations, obtained with SLHMC and DFT-MD simulations. The MSD is defined as $\langle r_k(t)^2 \rangle = \frac{1}{N_{\text{Si}}} \sum_{k=1}^{N_{\text{Si}}} (r_k(t) - r_k(0))^2$, where $N_{\text{Si}}$ is the number of Si atoms and $r_k(t)$ is the position of $k$-th atom relative to the center-of-mass of the system at time $t$. These MSD curves, which should converge to a constant value for large $t$ in solid states, indicate the length scale of the autocorrelation of atomic displacement. As expected, the MSD converges faster as $\Delta t_{\text{DFT}}$ increases, since the ANN-MD trajectory is longer. Assuming that the DFT calculations are the bottleneck of the SLHMC simulations, which was mostly the case
in our computations, SLHMC simulations become more efficient as the MSD converges. This is not only because the sampling of statistics becomes more efficient, but also because the training data becomes less correlated. When $\Delta t_{\text{DFT}}$ is long enough to be uncorrelated in a single MC step, the efficiency of SLHMC should become proportional to the acceptance ratio, which is eventually reflected by the difference between the ANN and DFT potentials and thus the quality of the ANN. In the present simulation, the acceptance ratio with the well-trained ANNs with $\Delta t_{\text{DFT}} = 50$ fs was around 40%, and the difference between the ANN and DFT potentials tended to 0.23 meV/atom on the average.

**Demonstration II: Dynamics of YNi$_2$B$_2$C.** The second test was on the dynamics of an unconventional phonon-mediated superconductor YNi$_2$B$_2$C ($T_c \sim 15$ K) [42-43]. Neutron scattering experiments have shown a strong temperature dependence of the phonon density of states (DOS) in this superconducting compound [46]. The temperature dependence arises from anharmonic vibrations that are not taken into account in static DFT calculations based on (quasi-)harmonic analysis at zero temperature [46, 47]. We show that a combination of the SLHMC and ANN-MD methods could be useful in this case. Since the SLHMC method optimizes the ANN potential in the configuration space of a given ensemble, the accuracy of ANN potential is guaranteed in ensuing ANN-MD simulations as the trajectories stay confined within this configuration space. Therefore, once the ANN potential is optimized in SLHMC, the ANN-MD runs should be stable for long times, which may not necessarily be the case for complex systems with several elements (in the present case, four elements) for the methods proposed earlier.

The SLHMC and ANN-MD simulations were carried out for a supercell containing 16 Y atoms, 32 Ni atoms, 32 B atoms, and 16 C atoms. The ANN and DFT simulations were setup in the same way as those in the previous section. The SLHMC simulations were at 1000 K with the step sizes $\Delta t_{\text{ANN}} = 0.25$ fs, $\Delta t_{\text{DFT}} = 2.5$ fs, and $\Delta t_T = 250$ fs. The difference of the trained ANN and DFT potentials was found to be about 0.4 meV/atom on average. The DOS was computed via the Fourier transform of velocity autocorrelations [48] from the ANN-MD simulations. For statistical convergence, ten Newtonian trajectories were run independently for 100 ps with the step size $\Delta t_{\text{MD}} = 1$ fs. These trajectories were restarted from the equilibrated configurations of NVT ensemble at 60 K and 300 K to reflect the temperature dependence of the DOS. As expected, all the ANN-MD trajectories were found to be stable for 100 ps.

As shown in Figure 5, the phonon density of states depends on temperature, which is consistent with the neutron scattering experiments [49]. This result confirms that anharmonic effects of phonons are important in this material. The crystal structure of YNi$_2$B$_2$C is similar to that of high-$T_c$ cuprates and strongly anisotropic superconducting pairing has been suggested [43-47]. Thus anharmonic effects of phonons appear to play a key role in this unconventional superconductor.

**Conclusions.** We proposed a new method called SLHMC to compute thermodynamic properties exactly based on DFT using an approximate ML potential that is trained on the fly to accelerate sampling. The ML potential is optimized automatically by using a training data set of a given ensemble that is generated exactly. The ML potential thus obtained can be used safely in ML-MD simulations to compute dynamic properties in thermal equilibrium, since the ML-MD simulations are stable for long times. Proof-of-concept calculations were presented for the thermodynamics of SiO$_2$ and the dynamics of YNi$_2$B$_2$C, which demonstrated the usefulness of SLHMC.

As can be expected from the acceptance probability in Eq. (5), the efficiency of SLHMC depends strongly on the balance between the system size and the qual-
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* nagni.yuki@jaea.go.jp
† shiga.motoyuki@jaea.go.jp

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