Application of machine learning potentials to predict grain boundary properties in fcc elemental metals

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Accurate interatomic potentials are in high demand for large-scale atomistic simulations of materials that are prohibitively expensive by density functional theory (DFT) calculation. In this study, we apply machine learning potentials in a recently constructed repository to the prediction of the grain boundary energy in face-centered-cubic elemental metals, i.e., Ag, Al, Au, Cu, Pd, and Pt. The systematic application of machine learning potentials shows that they enable us to predict grain boundary structures and their energies accurately. The grain boundary energies predicted by the MLPs are in agreement with those calculated by DFT, although no grain boundary structures were included in training datasets of the present MLPs.

I. INTRODUCTION

Grain boundaries are interfaces between differently oriented crystals of the same phase [1]. The microstructures of grain boundaries can affect various properties of polycrystalline materials, including mechanical, thermal, and electrical properties [2–5]. Thus, an attractive topic in materials science has been to establish the relationship between the properties of crystalline materials and grain boundary structures. Many theoretical studies have been made to cover a broad range of grain boundary structures and their excessive energies. Early fundamental studies employed pair potentials, such as the Lennard–Jones and Morse forms, to investigate the generic properties of grain boundaries such as the presence of cusps in a map of the rotation angle and the grain boundary energy [6–8]. Empirical interatomic potentials such as the Finnis–Sinclair (FS) potentials [9] and embedded atom method (EAM) [10] potentials have been widely used to investigate symmetric and asymmetric grain boundaries of metallic materials. Quantitative predictions are becoming possible [11–20], and strong correlations between theoretical and experimental grain boundary energies have been shown, especially for grain boundaries in elemental Al and Ni, which exhibit low grain boundary energies [21, 22]. However, the prediction error in the grain boundary energy may be significant in grain boundaries showing higher grain boundary energies. This error originates from the fact that their microscopic grain boundary structures differ from the atomic environment used to estimate interatomic potentials.

Density functional theory (DFT) calculation [23, 24] is an alternative way to predict grain boundary properties accurately. However, DFT calculation is practically impossible to apply to large-scale models of grain boundaries owing to its computational cost. Therefore, interatomic potentials that enable us to predict grain boundary properties accurately have been in high demand. Over the last decade, many groups have proposed frameworks to develop machine learning potentials (MLPs) based on extensive datasets generated by DFT calculation [25–46]. The MLPs significantly improve the accuracy and transferability of interatomic potentials. Also, MLPs themselves are becoming available, such as those in MACHINE LEARNING POTENTIAL REPOSITORY [47] developed by one author of this paper.

In this paper, we demonstrate the predictive power of MLPs in the MLP repository for grain boundary properties. We systematically evaluate the structures and excessive energies of (100) symmetric tilt grain boundaries (STGBs), ⟨110⟩ STGBs, and ⟨100⟩ pure-twist grain boundaries in the face-centered-cubic (fcc) elemental metals of Ag, Al, Au, Cu, Pd, and Pt. They are compared with those obtained from EAM potentials and DFT calculations. The MLP repository contains a set of Pareto optimal MLPs with different trade-offs between accuracy and computational efficiency; hence, we carefully determine appropriate MLPs to predict grain boundary properties.

II. METHODOLOGY

A. Modeling and structure optimization of grain boundaries

Macroscopic structures of grain boundaries are characterized by five geometrical degrees of freedom. We choose three variables to specify the direction of the rotation axis and the rotation angle, which describe the misorientation between crystal lattices, and two variables to specify the direction of the boundary plane normal [1]. For a given set of macroscopic variables, the microscopic structure is associated with three degrees of freedom regarding rigid body displacements: two components parallel to the boundary plane and one component normal to the
plane. Hence, the globally optimal microscopic structure for a given set of macroscopic variables is achieved by optimizing the three microscopic variables in terms of potential energy.

In this study, we investigate only STGBs and pure-twist grain boundaries. The periodicity of an STGB is identified from the orthogonal projection of its coincident site lattice (CSL) to its boundary plane. Also, the periodicity of a pure-twist grain boundary is given by the orthogonal projection of its displacement shift complete (DSC) lattice to its boundary plane. Therefore, we restrict the ranges of the two in-plane microscopic variables to a domain defined by the periodicity of the grain boundaries.

We explore the globally optimal microscopic structure for a set of macroscopic variables using a multi-start method. The multi-start method involves local structure optimizations for a given set of initial structures and regards the structure with the lowest energy among the converged final structures as the globally optimal structure. We use the conjugate gradient method implemented in the LAMMPS code [48] for the local structure optimizations. Initial microscopic structures are introduced from a $4 \times 4$ grid for the two in-plane components and a sequence for the component normal is introduced to the boundary plane. For each initial microscopic structure, a calculation model is generated using PYMATGEN [49]. This model contains two parallel boundaries perpendicular to the c-axis of the model, separated by fcc layers corresponding to four repetitions of a cell of the CSL. However, the local structure optimization starting from some of the initial microscopic structures fails to converge when using both the MLPs and the EAM potentials, as shown in the next section. These structures are ignored in finding the globally optimal microscopic structure. Note that the optimization of the microscopic structure is performed in the whole domain here, although it is more efficient to restrict the domain to its symmetrically nonequivalent domain.

### B. Machine learning potentials

We employ MLPs in MACHINE LEARNING POTENTIAL REPOSITORY [47] developed by one author of this paper to obtain the globally optimal microscopic structures of STGBs and pure-twist grain boundaries. In the repository, a set of Pareto optimal MLPs with different trade-offs between accuracy and computational efficiency is available, from which one can choose an appropriate MLP in accordance with the target and purpose. Potential energy models of the MLPs are either a polynomial model of Gaussian-type pairwise structural features or a polynomial model of polynomial invariants for the $O(3)$ group, which are derived by a group-theoretical approach [50].

The Pareto optimal MLPs in the repository have been developed using a dataset generated from structure generators. For Ag, Al, Au, and Cu, we adopt the Pareto optimal MLPs developed from a structure generator set composed of the fcc, body-centered-cubic (bcc), hexagonal-close-packed (hcp), simple cubic (sc), $\omega$, and $\beta$ tin structures. The dataset is composed of 3,000 structures constructed by introducing random lattice expansion, random lattice distortion, and random atomic displacements into a supercell of the equilibrium structure for one of the structure generators. For Pd and Pt, we employ another set of 82 prototype structures as the structure generator set because the dataset derived from the six structure generators is not available in the repository. The dataset consists of 10,000 structures generated by the same procedure as above. For all structures in the dataset, DFT calculations were performed using the plane-wave-basis projector augmented wave method [51] within the Perdew–Burke–Ernzerhof exchange-correlation functional [52] as implemented in the VASP code [53–55]. Note that the datasets contain no structures generated from grain boundary models.

### III. RESULTS AND DISCUSSION

First, we systematically calculate the grain boundary energies of five grain boundaries using the whole set of Pareto optimal MLPs for each elemental metal. They are the $\Sigma 5$ (100) STGB (at 53.1 degrees), the $\Sigma 3$ (110) STGB (at 70.5 degrees), the $\Sigma 9$ (110) STGB (at 109.5 degrees), the $\Sigma 3$ (110) pure-twist grain boundary (at 38.9 degrees), and the $\Sigma 5$ (100) pure-twist grain boundary (at 36.9 degrees), the calculation models for which can be represented by a small number of atoms. Then, we find an accurate MLP requiring only a reasonable computational time to investigate the whole set of grain boundaries.

Figure 1 shows the convergence behavior of the grain boundary energy in terms of the computational time, obtained using the whole set of Pareto optimal MLPs. The grain boundary energy is identical to the lowest energy among the grain boundary energies of the microscopic structures. The grain boundary energy of a microscopic structure is measured from the energy of the equilibrium fcc structure. The computational time corresponding to the model complexity of an MLP is the elapsed time normalized by the number of atoms for a single point calculation of the energy, the forces, and the stress tensors [56]. As can be seen in Figure 1, the grain boundary energy converges well in all of the elemental metals and grain boundaries. Consequently, successive calculations for the whole set of grain boundaries are performed using the MLP that requires the lowest computational time among the MLPs showing convergence.

We also examine the transferability of the MLPs to the prediction of the grain boundary structures and energies because the datasets used in developing the MLPs contain no grain boundary structures. Therefore, we evaluate the grain boundary energies of the $\Sigma 3$ (110) STGB (at 70.5 degrees), the $\Sigma 3$ (110) STGB (at 109.5
Figure 1. Grain boundary energies of $\Sigma 5$ (100) STGB in 53.1 degrees, $\Sigma 3$ (110) STGB in 70.5 degrees, $\Sigma 3$ (110) STGB in 109.5 degrees, $\Sigma 9$ (110) STGB in 38.9 degrees, and $\Sigma 5$ (100) pure-twist grain boundary in 36.9 degrees for elemental Ag, Al, Au, Cu, Pd, and Pt, predicted using the Pareto optimal MLPs. The grain boundary energies computed by DFT calculation are also shown by broken lines.
degrees), the $\Sigma^9$ (110) STGB (at 38.9 degrees), the $\Sigma^5$ (100) STGB (at 53.1 degrees), and the $\Sigma^5$ (100) pure-twist grain boundary (at 36.9 degrees) by DFT calculation, and compare them with those predicted using the MLPs. Figure 1 shows the DFT values of the grain boundary energy only for the grain boundary structures, DFT calculations for which converge successfully. They are close to the grain boundary energies of the selected MLPs. Therefore, the selected MLPs should have high predictive power for grain boundary structures and their energies even though no grain boundary structures were used to develop the MLPs.

Table I lists the model parameters of the selected MLPs. Fast MLPs are selected for Ag and Cu, while computationally expensive MLPs are selected for the others. Table I also shows the prediction errors for the datasets used in developing the MLPs. The MLPs for Pd and Pt show significant prediction errors, which originate from the fact that the datasets contain many hypothetical structures such as the graphite-type structure. Although the selected MLPs exhibit significant prediction errors for such abnormal structures, they show much smaller prediction errors for typical metallic structures, including grain boundary structures, as shown above.

After confirming the transferability of the MLPs, we calculate the energies of the grain boundary structures: (100) STGBs ($\Sigma^5$, $\Sigma^{13}$, $\Sigma^{17}$, $\Sigma^{25}$, $\Sigma^{29}$, $\Sigma^{41}$), (110) STGBs ($\Sigma^3$, $\Sigma^9$, $\Sigma^{11}$, $\Sigma^{17}$, $\Sigma^{19}$, $\Sigma^{27}$, $\Sigma^{33}$, $\Sigma^{41}$, $\Sigma^{43}$), and (100) pure-twist grain boundaries ($\Sigma^5$, $\Sigma^{13}$, $\Sigma^{17}$, $\Sigma^{25}$, $\Sigma^{29}$, $\Sigma^{37}$, $\Sigma^{41}$). Most of them are represented by large-scale models, hence they cannot be calculated by DFT calculation because of the large computational resources required. Figure 2 shows the rotation angle dependence of the grain boundary energy obtained using the MLPs and EAM potentials [57–61]. The values of the grain boundary energy in Al, Cu, and Pd computed using the MLPs are consistent with those computed using the EAM potentials and those computed by DFT calculation. Therefore, the MLPs and the EAM potentials have high predictive power for the grain boundary structures and their energies. In Ag, Au, and Pt, the values of the grain boundary energy computed using the MLPs are almost the same as those computed by DFT calculation, whereas they deviate from those computed using the EAM potentials. The MLPs should be more reliable than the EAM potentials for obtaining not only the grain boundary structures and their energies but also the other defect structures in Ag, Au, and Pt. Note that a fine sequence is required for the component normal to the boundary plane to obtain converged microscopic structures when using the EAM potentials for Ag and Au. This implies that the EAM potentials for Ag and Au lack accuracy for predicting the potential energy surface around the globally optimal microscopic structure.

IV. CONCLUSION

We have examined the predictive power of MLPs in an MLP repository for grain boundary properties by systematically evaluating the grain boundary energy for (100) STGBs, (110) STGBs, and (100) pure-twist grain boundaries in the fcc elemental metals of Ag, Al, Au, Cu, Pd, and Pt. In every elemental metal, the values of the grain boundary energy computed using the MLP are consistent with those computed by DFT calculation. We emphasize that the training datasets used to develop the MLPs contain no grain boundary structures. Therefore, the consistency indicates that the MLPs have high predictive power for the grain boundary structures and energies. The present results also imply that the MLPs in the repository, including those for other systems, should be useful in accurately predicting grain boundary properties and other complex defect properties.

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The grain boundary energies computed by DFT calculation are also shown by crosses.

For comparison, the grain boundary energies predicted using EAM potentials for Ag [57, 58], Al [59], Au [57, 60], Cu [61], Pd [60], and Pt [60] are shown by open symbols. The grain boundary energies computed by DFT calculation are also shown by crosses.
Table I. Model parameters of the MLPs used to estimate the grain boundary structures and energies. The identification of the feature type, the model type, and the polynomial orders can be found in Ref. [47].

| MLP-ID    | Ag  | Al  | Au  | Cu  | Pd  | Pt  |
|-----------|-----|-----|-----|-----|-----|-----|
| RMSE (energy) [meV/atom] | pair-44 | 2.2 | 0.8 | 0.7 | 2.2 | 6.3 |
| RMSE (force) [eV/Å] | | 0.010 | 0.008 | 0.012 | 0.013 | 0.097 | 0.172 |
| Time [ns/atom/step] | 0.05 | 1.85 | 0.66 | 0.04 | 0.52 | 0.63 |
| Number of coefficients | 815 | 1100 | 475 | 285 | 500 | 1595 |
| Feature type | Pair | Invariants | Invariants | Pair | Invariants | Invariants |
| Cutoff radius [Å] | − | 7.0 | 8.0 | 6.0 | 7.0 | 6.0 |
| Number of radial functions | − | 15 | 15 | 10 | 10 | 5 |
| Model type | 2 | 2 | 2 | 2 | 2 | 2 |
| Polynomial order (function $F$) | 3 | 3 | 3 | 3 | 3 | 3 |
| Polynomial order (invariants) | − | − | − | − | − | − |
| Spherical harmonics truncation $\{l_{\text{max}}, l_{\text{max}}\}$ | − | [4, 4] | [4, 4] | − | [4, 0] | [4, 2] |

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