Efficient moment tensor machine-learning interatomic potential for accurate description of defects in Ni-Al Alloys

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Abstract

Combining the efficiency of semi-empirical potentials with the accuracy of quantum mechanical methods, machine-learning interatomic potentials (MLIPs) have significantly advanced atomistic modeling in computational materials science and chemistry. This necessitates the continual development of MLIP models with improved accuracy and efficiency, which enable long-time scale molecular dynamics simulations to unveil the intricate underlying mechanisms that would otherwise remain elusive. Among various existing MLIP models, the moment tensor potential (MTP) model employs a highly descriptive rotationally-covariant moment tensor to describe the local atomic environment, enabling the use of even linear regression for model fitting. Although the current MTP model has achieved state-of-the-art efficiency for similar accuracy, there is still room for optimizing the contraction process of moment tensors. In this work, we propose an effective genetic algorithm based optimization scheme that can significantly reduce the number of independent moment tensor components and intermediate tensor components. This leads to a speedup of nearly one order of magnitude in efficiency and also improved accuracy compared to the traditional MTP model for intricate basis sets. We have applied our improved MTP model to predicting the energetic and dynamical properties of various point and planar defects in Ni-Al alloys, showing overall good performances and in general outperforming the semi-empirical potentials. This work paves the way for fast and accurate atomistic modeling of complex systems and provides a useful tool for modeling defects in Ni-Al alloys.

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I. INTRODUCTION

Nickel-based high-temperature alloys hold significant importance in engineering applications, particularly in gas turbines and aerospace, owing to their exceptional high-temperature mechanical properties. The superior mechanical properties originate from the ordered γ' -Ni₃Al phase embedded in the γ phase matrix [1]. The presence of a lattice misfit leads to the formation of a dislocation network between the γ and γ' phases, which prevent dislocations sliding through the γ' phase, resulting in increased strength and creep resistance [2]. Moreover, defects such as impurities, vacancies, dislocations, and stacking faults are prevalent in these alloys, significantly influencing their mechanical strength and resistance to creep. For instance, manipulating complex stacking faults and twinning boundaries has been discovered to effectively enhance the hardening properties of Ni-Al alloys [3]. Furthermore, the aggregation of vacancies in Ni-Al alloys can lead to the formation of vacancy-type dislocation loops, voids, and stacking fault tetrahedra (SFT) under conditions like quenching, irradiation, plastic deformation, and exposure to a hydrogen environment [4–7]. These defects have a direct impact on the mechanical properties of the alloys. Therefore, accurately predicting the energetic and dynamic properties of these defects in Ni-Al alloys is crucial for optimizing their mechanical performance.

Density functional theory (DFT) has significantly advanced our comprehension of defect behavior in the Ni-Al alloys [8–23]. For instance, Siegel [8] reported that alloying elements can reduce the stacking fault energies in Ni. Shang *et al.* [9] further studied the effects of various alloying elements on stacking fault energies in Ni, demonstrating that most alloying elements decreased the stacking fault energy, with a more pronounced effect observed when the alloying element is further from Ni in the periodic table. Zhu *et al.* [18] highlighted the significant strengthening effects of the alloying elements Re, W, and Mo on the Ni and Ni₃Al systems, which increased the slip energy barriers of the trailing slip process in the γ phase and the leading slip process in the γ' phase. Using a DFT based flexible boundary condition approach, Tan *et al.* [19] investigated the core structure of $\frac{1}{2}$ [110] screw dislocation in the γ phase and [110] screw super dislocation in the γ' phase, determining the split distances of planer faults in the dislocation core structure, in good agreement with experimental observations. Zhao *et al.* [20] investigated the stability of vacancy clusters in Ni using *ab initio* molecular dynamics (AIMD), revealing that the SFT was the most thermodynamically stable configuration for large vacancy clusters in Ni.

Despite the extensive DFT investigations on the Ni-Al alloys, accurately modeling defects re-

mains a challenging task. On the one hand, a large simulation cell is often required to eliminate the artificial interactions between the defect and its periodic replicas, thereby substantially increasing the computational expenses. This necessity becomes especially pronounced for intricate defects like vacancy clusters, dislocations, and stacking faults. On the other hand, capturing the dynamical properties of defects necessitates a long timescale that is usually beyond the reach of AIMD simulations. To overcome the limitations of DFT, several semi-empirical interatomic potentials have been developed for the Ni-Al binary system. Among these potentials, the most successful ones for Ni are the embedded atom method (EAM) potential developed by Voter *et al.* in 1986 [24], Mishin *et al.* in 2004 [25] and 2009 [26], and the EAM potential optimized for NiAl and Ni₃Al systems developed by Wang et al. [27]. Besides, Kumar et al. [28] developed a third-generation charge-optimized many-body potential, which was featured by its accurate description of planer defects within the γ and γ' phase as well as their interfaces with Al₂O₃. Notably, Avik *et al.* [29] recently developed a modified EAM potential for the Ni-Al system that worked in a wide temperature range and can accurately reproduce experimental melting points across all compositions. Although these physics-based semi-empirical potentials were capable of reproducing the target properties used in the fitting such as experimental lattice parameters, elastic constants, cohesive energy, and vacancy formation energy, they often suffered from the limited accuracy and failed to capture the properties of complex defects over a large phase space, because of their simple and inflexible analytical forms.

Machine-learning interatomic potentials (MLIPs) have emerged as an elegant solution to bridge the expensive DFT calculations and less accurate semi-empirical potentials [30–63]. These MLIPs utilize machine learning techniques to fit the high-fidelity quantum mechanical data, thereby maintaining the high efficiency of semi-empirical potentials while preserving quantum mechanical accuracy. In recent years, the MLIPs have been successfully applied to metals and alloys [64–81]. With variations in the construction of the training set, structural descriptors, and regression techniques, several MLIP models have been proposed. Behler and Parrinello [30] pioneered the locality assumption and proposed a high-dimensional neural network potential (HDNNP) model. As of now, the HDNNP model has advanced to the fourth generation, incorporating long-range Coulomb interactions, charge transfer, and magnetic moments [32, 82]. The Deep Potential model [33, 34] also employed a deep neural network but using an embedding network of atomic coordinates as local configuration descriptors, and have achieved great success in various systems [79, 83–86]. Another popular and successful MLIP model is the kernel-based Gaussian Approximation Potential (GAP) proposed by Bartók *et al.* [37, 38], which employed the descriptor of the Smooth Overlap of Atomic Positions (SOAP) and the Gaussian process regression [37, 38], demonstrating promising results in many applications [51]. Jinnouchi *et al.* [39, 40] further developed the GAP model by incorporating an on-the-fly active learning procedure within AIMD simulations using the Bayesian force errors, effectively addressing the laborious process of conventional training set construction. Despite the popularity of the GAP potential, the kernel-based models suffer efficiency issues, especially as the training set grows large. Instead of employing complex neural networks or kernel functions, linearly parameterized models such as the Spectral Neighbor Analysis Potential (SNAP) [43], Moment Tensor Potential (MTP) [44, 45], and Atomic Cluster Expansion (ACE) [46–48] have been developed owing to their highly descriptive nature. Zuo *et al.* [87] systematically assessed the accuracy and efficiency of the HDNNP, GAP, MTP, SNAP and quadratic SNAP (qSNAP) [88] models on various lattice types, demonstrating that among these tested models, the MTP potential exhibits the highest efficiency for the same level of accuracy.

While the MTP model has reached state-of-the-art levels in efficiency and accuracy, there remains room for improvement. In this work, we advanced the MTP model by developing an elegant optimization scheme for the contraction of moment tensors. Thanks to the utilization of lower-rank moment tensors and more efficient contraction rules, the number of independent moment tensor components and intermediate tensor components in our improved MTP model was significantly reduced. This resulted in a nearly tenfold increase in speed and enhanced accuracy as compared to the traditional MTP model for the basis sets with high level of complexity. As a benchmark application of our improved MTP model, we have systematically assessed its performance in describing the energetic and dynamical properties of various point and planar defects in Ni-Al alloys, an established challenging task for MLIPs. Our results showcased consistently good performance of our developed MTP model, in general outperforming the semi-empirical EAM potentials. This was manifested by accurately predicting various properties such as the lattice constants, elastic properties, energy-volume curves, and phonon dispersions as well as the melting points of bulk Ni and Ni₃Al. Furthermore, our MTP model accurately predicted the formation and binding energies of vacancy clusters, antisite defects, stacking faults, and the diffusion behavior of vacancy clusters. This work paves the way for fast and accurate atomistic modeling of complex systems and lays foundation for further improving the current MTP model for Ni-Al alloys by expanding the phase space of the training set.

The paper is organized as follows. In Sec. II we will detail the methodology of our improved

MTP model. Particular emphasis is devoted to the proposed optimization scheme for contracting moment tensors and basis set selection. Technical details and computational setups will be provided in Sec. III. The extensive results will be presented and discussed in Sec. IV, followed by the conclusion in Sec. V.

II. METHOD DEVELOPMENT

A. Moment tensor potential

We start from the introduction of the moment tensor potential (MTP) proposed by Shapeev [44]. The key quantity of the MTP is the rotationally covariant moment tensor descriptor [44]

$$M_{\mu,\nu}(\mathfrak{n}_i) = \sum_j f_{\mu,\nu}\left(\left|r_{ij}\right|, z_i, z_j\right) f_c(\left|r_{ij}\right|) \underbrace{\mathbf{r}_{ij} \otimes \ldots \otimes \mathbf{r}_{ij}}_{\nu \text{ times}}, \qquad (1)$$

which describes the local atomic environment n_i of atom *i*. The moment tensor $M_{\mu,\nu}$ is a tensor of rank ν . $f_{\mu,\nu}$ denotes radial basis functions (e.g., Chebyshev polynomials), z_i denotes the species index of atom *i*, and f_c represents the cutoff function function that approaches to zero smoothly at a specific cutoff radius r_c . \mathbf{r}_{ij} represents the distance vector from atom *i* to atom *j*, and the symbol \otimes denotes the Kronecker product.

It is worth noting that the rank of the moment tensor corresponds to the rank of the momenta of the weighted atom mass density [44]. Physically, one can think of the moment tensor descriptors as analogues to mass distribution properties. For instance, the rank-0 moment tensor descriptor simply gives the total mass and the rank-1 moment tensor descriptor describes the center of mass. The rank-2 moment tensor descriptor represents the second moments of inertia, while the rank-3 and rank-4 moment tensor descriptors capture the skewness and kurtosis of the mass distribution, respectively.

By appropriately contracting a set of k moment tensors M_{μ_i,ν_i} , one is able to obtain a scalarvalued k-body function [44]. Such contraction can be represented by a symmetric index matrix α of size $k \times k$ with diagonal terms being zero. The *i*-th row of the matrix α corresponds to the moment tensor M_{μ_i,ν_i} , and the sum of the elements in the *i*-th row of the matrix α is equal to ν_i , (i.e., $\nu_i = \sum_j \alpha_{ij}$). The off-diagonal element α_{ij} describes how many dimensions are contracted between the moment tensors M_{μ_i,ν_i} and M_{μ_j,ν_j} . Using this contraction representation, the scalarvalued function can be described as $B_{\alpha} = \prod_i \alpha M_{\mu_i,\nu_i}$ [44]. To better illustrate the contraction, here we present an example. Suppose that we have three moment tensors $M_{1,3}$, $M_{2,2}$ and $M_{3,1}$ [i.e., $\mu = (1, 2, 3)^T$ and $\nu = (3, 2, 1)^T$] and a contraction rule $\alpha = \begin{pmatrix} 0 & 2 & 1 \\ 2 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}$, then the final contracted

scalar function B_{α} can be represented in Einstein summation rule as $B_{\alpha} = (M_{1,3})_{ijk}(M_{2,2})_{ij}(M_{3,1})_k$.

The scalar basis functions $\{B_{\alpha}\}$ are invariant under translation, rotation and reflection. It has been shown by Shapeev [44] that all these scalar functions form a complete basis set. Therefore, the local atomic energy U_i of atom *i* can be written as linear combinations of these scalar basis functions

$$U_i = \sum_{\theta} \xi_{\theta} B_{\theta}, \tag{2}$$

where ξ_{θ} are linear coefficients. The summation over all atoms then yields the potential energy of the system $U = \sum_{i} U_{i}$.

B. Optimization of moment tensors contraction

In practical implantation, directly computing the contractions is computationally demanding, since the entries to be summed grow exponentially with the count of contracted indices (namely, 3^m when contracting *m* indices between two moment tensors). Thanks to the symmetric nature of the moment tensors, this computational cost can be reduced to (m + 2)(m + 1)/2 when incorporating symmetry of the moment tensors. To reduce the cost further, Shapeev [44] found an ingenious scheme. In this scheme, as illustrated in Fig. 1, a contraction process can be represented as a tree. Starting from a scalar function which is defined by contractions of moment tensors, a tensor is decomposed into contraction of two intermediate tensors iteratively until moment tensors are reached. This process actually defines the order of contractions and represents the intermediate results. It can be seen that decomposition for different scalar functions may contain the same intermediate tensors. Therefore, if all these common intermediate tensors are computed once and used for all, the entire tensors contraction cost can be further reduced.



Figure 1. Illustration of the tree representation of scalar decomposition. B_{α} and B_{β} are two different scalar functions, B_{η_i} are indeterminate tensors, and M_i are moment tensors. Note the common indeterminate tensors B_{η_2} and B_{η_4} between the two different scalar functions.

To illustrate this procedure in detail, the indices of a tensor B_{α} that is decomposed into a tensor B_{γ} of rank k_{γ} and a tensor B_{η} of rank k_{η} can be expressed as

$$\alpha = \begin{pmatrix} \gamma' & \phi \\ \phi^T & \eta' \end{pmatrix},\tag{3}$$

$$\gamma'_{i,i} = \gamma_{i,i} - \sum_{j} \phi_{i,j}, \quad \eta'_{i,i} = \eta_{i,i} - \sum_{j} \phi^{T}_{i,j},$$
 (4)

$$\gamma'_{i,j} = \gamma_{i,j}, \quad \eta'_{i,j} = \eta_{i,j}, \quad (i \neq j), \tag{5}$$

where γ' and η' are the diagonal blocks of the index matrix α with sizes of $k_{\gamma} \times k_{\gamma}$ and $k_{\eta} \times k_{\eta}$, respectively, and ϕ is the off-diagonal block of α with size $k_{\gamma} \times k_{\eta}$, which defines contractions between B_{γ} and B_{η} . The non-zero diagonal terms α_{ii} of non-scalar index α denote that the α_{ii} indices of moment tensor M_i are not involved in contraction, whereas the off-diagonal terms α_{ij} represent contracting α_{ij} indices between moment tensors M_i and M_j . The indices α , γ , η and ϕ define a contraction rule to obtain B_{α} from B_{γ} and B_{η} .

Based on above analysis, the computation routine for the moment tensors contraction in practice can then be established. First, the components $\{m_{i,j}\}$ of moment tensor M_i can be calculated directly from atomic coordinates. The components of intermediate tensor B_{η_3} in Fig. 1, for example, can be calculated by $b_{\eta_3,i} = \sum_k m_{3,k} m_{4,k}$ where k loops over contracted indices, and so do other intermediate tensors. Taking B_i as root node and B_i and B_m as leaf nodes for instance, the components of B_i can be expressed as $b_{i,j} = \sum_k b_{l,k} b_{m,k}$. By assigning each nonequivalent tensor components with an unique ID, each intermediate tensor component and scalar tensor component can be evaluated through a collection of tuples $\{(n, d_i, k_i, l_i)\}$

$$b_n = \sum_i d_i \cdot b_{k_i} \cdot b_{l_i},\tag{6}$$

where d_i is the degeneracy due to tensor symmetry, n, k_i and l_i are unique IDs of tensor components. We refer to these tuples as *times rules*. Once these rules have been built, one can employ them at any time without rebuilding them.

It needs to be noted that for a specific scalar function B_{α} , the decomposition routine is not unique. In fact, for a collection of scalar functions $\{B_{\alpha}\}$, different choices of decomposition routines for each scalar function can lead to varied numbers of nonequivalent tensor components and times rules. This, in turn, significantly increases the computational cost of the model training and applications. Due to the large number of possible decomposition routines, it is practically impossible to loop over all cases. For instance, in the case of the moment tensors with level=28 as defined by Shapeev [44], there are more than 10^{2195} possible decomposition routines. Shapeev [44] simply pointed out that minimizing the sum of dimensions for each decomposition and maximizing tensor reuse can be served as a guideline for the selection of decomposition routines. Although this scheme may result in a reasonable solution, it could introduce ambiguity, e.g., when multiple decomposition routines have the same sum of dimensions or intermediate tensors, and thus might not consistently ensure the optimal solution.

To tackle this issue, we propose a straightforward approach to obtain the optimal decomposition routine for each scalar function using genetic algorithms (GA). Specifically, we construct all possible decomposition trees for each scalar function. These trees constitute a "tree collection" and are denoted as $\{T_i^{\alpha}\}$ for a scalar function B_{α} . For each scalar function B_{α} , we assign a "gene" k, which picks the k-th tree out of $\{T_i^{\alpha}\}$. For a collection of scalar functions, a sequence $\{k_1, k_2, \dots, k_n\}$ forms an individual, which represents a decomposition routine. The objective of optimizing the computation routine amounts to minimizing the number of nonequivalent tensor components and the count of times rules. To quantify this, we define a cost function F(R)

$$F(R) = m + 3t,\tag{7}$$

where R represents a specific routine, m is the number of nonequivalent tensor components, and t is the count of times rules. Adding more weight (i.e., "3") to the count of times rules is due to the

fact that this part is computationally more costly for relatively large number of basis sets. With the definition of F(R), the fitness function of each individual can then be defined as

$$f(R) = 1 - \frac{1}{1 + e^{-k \left(F(R) - F(R)_{\min}\right) + c}}.$$
(8)

Here, $F(R)_{min}$ represents the current minimum of F(R) and it will be changed dynamically during the optimization process. *k* and *c* are positive constants, which should be fine-tuned to prevent populations from being dominated by identical solutions. The detailed workflow can be described as follows:

- (1) Generate all nonequivalent scalar functions according to given conditions (i.e., maximum "level", order of many-body interactions, maximum coordination power, etc.) and then sort these scalar functions by the count of moment tensors involved in contraction.
- (2) Build tree collections for all scalar functions.
- (3) Generate initial populations of individuals. In this step, a score is assigned to each tree in the collection based on specific criteria. For example, a tree may gain ten points if it has the minimum sum of dimensions among all possible decompositions, with one point for each reuse of intermediate tensor and an additional point if a tensor can be decomposed into two identical tensors. Those trees with highest scores are selected as the initial guesses for the decomposition routine.
- (4) Generate child populations via mutations and cross-over operations. The mutation operator randomly selects a scalar function and replaces its current tree with another one from the collection, and the cross-over operator substitutes a subset of an individual's trees with those from another individual.
- (5) Calculate the fitness function of each individual and decide which individuals will be survived according to their fitness.
- (6) Repeat steps (4) and (5) until either the designated maximum number of iterations has been attained or the convergence criteria have been reached.

It should be mentioned that during the search process, we enforce that each tensor is assigned by one decomposition routine. This means that the genes representing the decomposition trees are not independent from each other. In other words, choosing a specific tree for a previouslyprocessed scalar function may render the tree structure invalid for a later-processed scalar function if they assign the same tensor with nonequivalent decompositions. This will introduce conflicts during the optimization process. To tackle this problem, when generating times rules for the laterprocessed tree, the branches forked from previously-processed tensors are chopped so that they can be consistent with the previously-processed tensors without changing the genes. After the optimization using the genetic algorithms, a greedy algorithm search is then carried out to further optimize the computation routine. In this step, all trees are looped from a low rank to a higher rank and the decomposition routine with the lowest loss function is eventually selected.

C. Basis set selection

After the rules of moment tensors contraction have been established, a subsequent question is which basis functions should be selected. Unlike the scheme proposed by Shapeev [44] which empirically defines the level of scalar basis as $lev(B_{\alpha}) = 4\mu + \nu + 1$ [see Eq. (1) for the meaning of μ and ν], here we redefine the level of scalar basis as $lev(B_{\alpha}) = 2\mu + \nu + 1$. We made such choice based on the consideration that the computational complexity of the MTP model primarily arises from two sources: (i) the calculation of the moment tensor components, and (ii) the computation of all tensor components through pre-generated contraction rules. To compute the moment tensor components, one must loop over all n_i neighboring atoms of a central atom *i*. The computational cost of this process exhibits a quadratic scaling with the moment tensor order ν and a cubic scaling with the radial cutoff r_c . This part of the computation can not be optimized and its cost may become dominant as v increases. Therefore, we limit the upper bound of v to a smaller value ($v \le 3$) to reduce the number of moment tensor components. Additionally, we retain the complexity of the basis functions by decreasing the penalty factor of μ from 4 to 2. Our tests showed that this scheme can greatly enhances the computational speed without compromising accuracy. To achieve higherorder polynomial terms, one can introduce scalar basis functions generated by the contraction of a larger number of moment tensors (representing higher-order many-body interactions).

III. COMPUTATIONAL DETAILS

All first-principles calculations and on-the-fly training set selection were performed using the Vienna *ab initio* simulation package (VASP) [89, 90]. The Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [91] within projected augmented wave method [92, 93] was employed. A plane-wave cutoff of 400 eV and a Monkhorst-Pack *k*-point grid with a spacing of ~0.2 Å⁻¹ were used, which ensure the convergence of total energy within 1 meV/atom. The convergence criteria for the electronic self-consistent calculation and structure optimization were set to 10^{-6} eV and 0.01 eV/Å, respectively.

The phonon dispersions were calculated by the frozen phonon method using the Phonopy code [94, 95]. For these calculations, a $3 \times 3 \times 3$ supercell and a finite displacement of 0.01 Å were employed.

All molecular dynamics simulations were conducted using the LAMMPS code [96] in the NPT ensemble at ambient pressure. The Langevin thermostat [97] and Parrinello-Raman method [98, 99] were used to control the temperature and pressure of the system. The time step was set to 2 fs.

IV. RESULTS AND DISCUSSION

A. Training and validation sets generation

The initial training structures were obtained using an on-the-fly learning procedure based on kernel-based Bayesian regression during AIMD simulations as implemented in the VASP code [39, 40]. The cutoff radius for the descriptors and the width of the Gaussian functions used for broadening the atomic distributions of the descriptors were set to 5 Å and 0.5 Å, respectively. The number of radial basis functions was set to 10 and 8 for the two-body and three-body descriptors, respectively. In order to generate a comprehensive training dataset, various initial structure models including bulk Ni and Ni₃Al, point defects such as Ni and Al vacancies, and defective Ni-Ni₃Al interfaces were built. Then, the on-the-fly AIMD samplings were performed by heating the structures from 700 to 1600 K at ambient pressure in an isothermal-isobaric ensemble using a Langevin thermostat [97] combined with the Parrinello-Rahman method [98, 99]. Throughout this process, a training dataset consisting of 6,092 configurations were obtained.

It is recognized that the kernel-based methods can encounter performance issues when training set grows large, resulting in large number of basis functions [87, 100]. To address this issue,

we refitted the on-the-fly generated dataset using a more efficient linearly parameterized MTP potential [44]. When building the MTP basis functions, the maximum number of moment tensors involved in contraction was set to 4, which resulted in up to five-body interactions considered. The maximum level of scalar functions was set to 26 and 8 radial basis functions were used.

Having obtained the initial MTP model that was refitted the on-the-fly generated dataset, we performed multiple active learning cycles based on Shapeev's generalized D-optimality criterion [45] to further sample the phase space and improve the transferability of the model. These cycles began by generating initial configurations that span a broad range of diversity, including various defects. Subsequently, we carried out 200 ps MD simulations and identified configurations with an extrapolation grade exceeding five. The DFT energies, forces and stress tensors of these selected configurations were then computed and incorporated into the training set at the end of each active learning cycle, followed by a refitting of the MTP model. This process was repeated until no configurations reached the given threshold of extrapolation grade . In this second-round active learning procedure, 2,358 new configurations were collected, leading to the eventual expansion of the training set to 8,450 configurations, on which the final MTP potential was trained.



Figure 2. **a**, **b**, **c**, Comparison of DFT and MTP predicted energies, forces and stresses, respectively. **d**, PCA analysis of training and validation datasets. Black and red points represent training and validation data, respectively.

To assess the accuracy of our developed MLIP model, a validation set consisting of 815 configurations was constructed using the same structure prototypes as the training set. The training and validation datasets were analyzed by principle component analysis (PCA) using the smooth overlap of atomic positions as local structure descriptors [101]. The PCA analysis, as shown in Fig. 2d, demonstrated that the validation set effectively covers the training set. Three distinct clusters were identified, which correspond to Ni-based, Ni₃Al-based and interface configurations. The Ni₃Al-based configurations spread in a larger area, which is due to the diffusion of vacancies and vacancy clusters leading to antisite defects and defect complexes.

B. Basis set selection and training of MLIP

Following the scheme we proposed in Sec. II B, here we aim to find out the optimal basis functions that yield both accurate and efficient MLIPs by examining five basis sets. Among them, three are the basis sets with levels of 18, 24, and 28, as defined in the scheme of Shapeev [44]. The remaining two are the basis sets generated using our proposed scheme, which are referred to as the 2653 and 4613 basis sets. The 2653 basis set considered up to five-body interactions and moment tensors of rank 4, leading to 2653 linear parameters, whereas the 4613 basis set accounted for up to seven-body interactions and moment tensors of rank 6, leading to 4613 linear parameters. For each basis set, 100,000 GA search iterations were performed. For all the basis sets, the parameter *c* in Eq. (8) was set to be 0.2 and the parameter *k* in Eq. (8) was set to 5×10^{-2} for the level-18 basis set, 5×10^{-3} for the level-24 and 2653 basis sets, and 5×10^{-4} for the level-28 basis set, respectively. The population size was set to be 40. At each iteration, 12 individuals were selected by roulette wheel selection to generate new individuals.

Table I presents the assessments of different basis sets on the accuracy and efficiency of resulting MLIPs. One can observe that the optimization process does not affect the accuracy, but it reduces the number of intermediate tensors, the number of nonequivalent tensor components, as well as the count of times rules for all the basis sets considered. This reduction becomes more pronounced for more complex basis sets. For example, for the simple basis set such as level-18, the reduction percentages are 8.0% for the number of intermediate tensors, 19.2% for the number of nonequivalent tensor components, and 19.0% for the count of times rules. By contrast, for more complex basis sets like level-28, which have more intricate decomposition routines, the optimization process exhibits more significant reductions in the numbers of intermediate tensors (18.2%), nonequivalent tensor components (41.3%) and times rules (43.9%).

Table I. Assessments of different basis sets on the accuracy and efficiency of MLIPs. The columns with "#Tensors", "#Components", and "#Times rules" represent the number of intermediate tensors, the number of nonequivalent tensor components, and the count of times rules, respectively. The columns with "Energies", "Forces", and "Stresses" indicate the training (upper row) and validation (lower row) RMSEs of energies (in meV/atom), forces (in meV/Å), and stress tensors (in kbar), respectively. MD times were measured on a $8 \times 8 \times 8$ supercell of bulk Ni₃Al (2048 atoms) with 100,000 time steps using 96 CPU cores and averaged for 5 independent runs. The column with "Efficiency" denotes the speedup of the MLIPs with optimized basis sets (OPT) as compared to those with non-optimized (INI) basis sets. Note that for these tests all the MLIPs were fitted on the same reduced training dataset (4800 structures) using a reduced cutoff radius of 5 Å.

Basis set	Į	#Tensors	#Components	#Times rules	Energies	Forces	Stresses	MD time (s)	Efficiency
18	INII	225	736	2799	3.64	0.045	2.22	151	1.00
	INI				2.40	0.042	1.52		
	OPT	205	595	2254	3.64	0.045	2.22	145	1.04
					2.40	0.042	1.52		
	INI	1222	4991	38970	3.00	0.035	1.52	917	1.00
24					1.98	0.033	1.11		
24	OPT	1038	3403	25732	3.00	0.035	1.52	711	1.29
					1.98	0.033	1.11		
	INI	3501	16504	186495	2.43	0.030	1.24	4418	1.00
20					1.53	0.028	0.97		
28	OPT	2863	9683	104664	2.43	0.030	1.24	2580	1.71
					1.53	0.028	0.97		
	INI	NI 2969	4764	15157	1.89	0.029	1.20	274	1.00
2(52					1.38	0.028	0.92		
2653	OPT	OPT 2854	3822	11581	1.89	0.029	1.20	241	1.14
					1.38	0.028	0.92		
4613	INI	II 5264	7676	18634	1.62	0.026	1.02	474	1.00
					1.11	0.026	0.88		
	OPT	OPT 4964	6140	13322	1.62	0.026	1.02	403	1.17
					1.11	0.026	0.88		

As for the efficiency, although the the optimization process reduces both the number of tensor components and times rules by 19% for the level-18 basis set, it only sightly decreases the MD time cost (3%). This is because in this case the computation cost is primarily driven by calculating moment tensor components rather than contractions. However, when it comes to the more complex basis sets such as level-28, the optimized MLIP exhibits a notable 71% acceleration. This is due to the fact that for complex basis containing higher order many-body interactions and more intricate contraction operations, loop over times rules and computing derivatives associated with tensor components turn to dominate the computing cost. As a result, in this case the optimization process of contractions can significantly enhance the overall computational efficiency.

Next, we turn to evaluating the performances of the 2653 and 4613 basis sets that were generated using our proposed scheme. As compared to the level-28 basis set (with 2445 scalar functions), the 2653 basis set contains a similar number of scalar functions and fitting coefficients. However, the trained MLIP using the 2653 basis set is nearly 11 times faster than using the level-28 basis set. Furthermore, the former exhibits a higher accuracy (see Table I). The enhanced efficiency of the 2653 basis set can be attributed to the utilization of lower-rank moment tensors and more efficient contraction rules. This results in a notable decrease in the number of independent moment tensor components (scaling quadratically with the tensor rank) and intermediate tensor components. It is worth mentioning that theoretically, the level-28 basis set is expected to achieve superior accuracy owing to its higher polynomial order (22 compared to 12 for the 2653 basis set excluding the radial function component). However, the comparatively reduced accuracy of the level-28 basis set, as evidenced in our findings, may be attributed to the heightened complexity of the basis set, which complicates the optimization process.

While the 4613 basis set offers the highest accuracy, in this study, the 2653 basis set is ultimately chosen for its notable efficiency and relatively high accuracy. With the 2653 basis set, the MLIP was refitted on the full training dataset (8,450 structures) and a larger cutoff of 5.4 Å. To achieve greater accuracy of the MLIP, we implemented a two-step final fitting procedure. The first step was to obtain suitable initial linear coefficients. This was achieved by the following processes. A minimal basis set was initially extracted from the full basis set, utilizing the same radial basis functions. This minimal basis set was utilized to fit the training set. Subsequently, the initial linear coefficients were obtained by conducting a linear optimization on the full basis set, inheriting the radial basis functions of the potential. The weights of energies, forces and stress tensors used in fitting were set to 1, 0.01 and 0.005, respectively. The second step involved optimizing the fitting parameters. This was executed using the L-BFGS algorithm with 5000 iterations, followed by a least square linear fit to further refine the linear parameters.

The accuracy of the ultimately fitted MLIP was assessed against the DFT calculations. Figure 2 illustrates the comparison between DFT and MTP-predicted energies, forces, and stresses on both the training and validation sets, demonstrating outstanding agreement. The root mean square errors (RMSEs) of the energies, forces and stresses for the training set are 2.24 meV/atom, 0.029 $eV/Å^2$ and 0.11 GPa, whereas for the validation set the RMSEs are 1.64 meV/atom, 0.028 $eV/Å^2$ and 0.09 GPa, respectively. To conclude this section, we have developed an efficient and accurate MTP model for the Ni-Al systems.

C. Assessment of MLIP for predicting fundamental physical properties

We further assessed the performance of our developed MLIP on various physical properties of Ni and Ni₃Al systems. For comparison purposes, we also employed the most widely used EAM-type semi-empirical interatomic potential for the Ni-Al alloy developed by Mishin [26]. We first computed the lattice parameters, elastic constants, bulk modules, shear modules and Yong's modules as well as Poison's ratio of both bulk Ni and Ni₃Al. The results are compared to the predictions from DFT and EAM methods as well as the experimental data, as summarized in Table II. One can see that the MTP-predicted lattice parameters accurately reproduce the DFT results. The underestimation in the lattice parameters compared to experimental data can be attributed to two factors: (*i*) The experimental data were obtained at room temperature, and (*ii*) more importantly, the current MLIP does not incorporate magnetic effects. Furthermore, the MTP reproduces well the elastic properties compared to the DFT results—Both are in good agreement with the experiential data. Regarding the elastic properties, the EAMs also demonstrate good performance. This outcome is not so surprising as the employed semi-empirical EAM potential was derived through fitting specifically to the elastic properties [26].

		a	C_{11}	C_{12}	C_{44}	В	G	Ε	v
Ni	DFT	3.511	257.53	171.26	117.70	200.02	78.73	208.79	0.326
	MTP	3.508	238.71	176.04	113.09	196.93	67.86	182.61	0.345
	EAM	3.520	241.34	150.85	127.34	181.01	84.15	218.57	0.299
	EXP	3.524 [102]	249 ± 4^a	155±7 ^a	114 ± 12^{a}	190 ± 13^{a}	78 ± 5^a	197±15 ^{<i>a</i>}	0.296 ± 0.029^{a}
Ni ₃ Al	DFT	3.562	237.56	156.40	125.80	182.86	80.27	210.08	0.309
	MTP	3.563	236.50	155.81	117.98	182.71	76.80	202.08	0.316
	EAM	3.533	237.32	166.38	130.16	190.03	77.61	204.94	0.320
	EXP	3.567[103]	230[104]	150[<mark>104</mark>]	131[<mark>104</mark>]				
			221[<mark>105</mark>]	146[<mark>105</mark>]	124[<mark>105</mark>]	171[<mark>105</mark>]	77.8[<mark>106</mark>]	203.1[106]	0.398[105]

Table II. Lattice constant *a* (in Å), elastic constants (C_{11} , C_{12} and C_{44} , in GPa), elastic moduli (bulk modulus *B*, shear modulus *G* and Young's modulus *E*, in GPa), and Poisson's ratio *v* of bulk Ni and Ni₃Al predicted by DFT, MTP and EAM methods. The available experimental (EXP) values are given for comparison.

^a Ref. [107]



Figure 3. **a**, **b**, The energy-volume curves of bulk Ni and Ni₃Al predicted by DFT, MTP, and EAM methods. **c**, **d** The phonon dispersion relationships and phonon density of states of bulk Ni and Ni₃Al predicted by DFT, MTP, and EAM methods.

Figure 3 displays the predicted energy-volume curves, phonon dispersion relationships and phonon density of states of bulk Ni and Ni₃Al by DFT, MTP, and EAM methods. In comparison to the EAM potential, the developed MTP potential accurately reproduces the DFT-calculated energy-volume curves across all the volume ranges under consideration. By contrast, the EAM potential diverges from the DFT outcomes as the system moves away from the equilibrium volume. Regarding the phonon properties, our developed MTP potential demonstrates exceptional alignment with the reference DFT results, once more surpassing the EAM potential.

D. Description of the energetics of point defects by MLIP

Since a crucial goal of our developed MLIP is to provide an accurate description of point defects, including vacancies and vacancy clusters, within the Ni-Al system, this section extensively evaluated the MLIP on various types of point defects, including mono-vacancies, di-vacancies, trivacancies, vacancy clusters, and antisite defects.

The vacancy formation energy for mono-vacancy and vacancy clusters in bulk Ni is defined as

$$E_f(nV_{\rm Ni}) = E(nV_{\rm Ni}) - (N-n)E_{\rm pristine}/N,$$
(9)

where $E(nV_{Ni})$ indicates the total energy of the defective system containing *n* vacancies V_{Ni} , $E_{pristine}$ stands for the total energy of the pristine bulk Ni system with *N* atoms. In bulk Ni₃Al and Ni-Ni₃Al interface systems, the vacancy formation energy for mono-vacancy and vacancy clusters is defined as [108]

$$E_f(n_{\rm Ni}V_{\rm Ni}, n_{\rm Al}V_{\rm Al}) = E_{defect} - E_{prefect} + n_{\rm Ni}E_{ref}^{\rm Ni} + n_{\rm Al}E_{ref}^{\rm Al} + n_{\rm Ni}\mu_{\rm Ni} + n_{\rm Al}\mu_{\rm Al},$$
(10)

where E_{defect} represents the total energy of the defective system with n_{Ni} Ni vacancies and n_{A1} Al vacancies, $E_{perfect}$ denotes the total energy of the defect-free system, while E_{ref}^{Ni} and E_{ref}^{A1} are the reference energies of bulk Ni and bulk Al in a face-centered cubic cell, respectively, and μ_{Ni} and μ_{A1} stand for the chemical potentials of the Ni and Al atoms in bulk Ni₃Al, respectively. Here, a Ni-rich reservoir was considered, meaning that $\mu_{Ni}=0$ eV and μ_{A1} is equal to the chemical potential of bulk Ni₃Al.

The binding energy of vacancy clusters is calculated as

$$E_b(n_{\rm Ni}V_{\rm Ni}, n_{\rm Al}V_{\rm Al}) = n_{\rm Ni}E_{V_{\rm Ni}} + n_{\rm Al}E_{V_{\rm Al}} - E_{defect} - (n_{\rm Ni} + n_{\rm Al} - 1)E_{prefect},$$
(11)

where E_{defect} represents the total energy of the defective system with n_{Ni} Ni vacancies and n_{Al} Al vacancies, $E_{perfect}$ denotes the total energy of the defect-free system, $E_{V_{Ni}}$ and $E_{V_{Al}}$ denote the total energies of the systems with a Ni vacancy and an Al vacancy, respectively. Following this convention, positive and negative binding energies denote the attractive and repulsive interactions of the vacancy clusters, respectively.

The antisite defect Ni_{Al} denotes a defective system in which a Ni atom occupies an Al site, while the antisite defect Al_{Ni} signifies a defective system where an Al atom occupies a Ni site. An antisite pair (Al_{Ni} , Ni_{Al}) is formed by exchanging the positions of a Ni and an Al atom. The formation energy of an antisite defect or an antisite pair is simply calculated as $E_f(\text{antisite}) = E_{defect} - E_{prefect}$, where E_{defect} and $E_{prefect}$ are the total energies of the systems with and without antisite defects, respectively.

Table III. Formation and binding energies of point defects and clusters in bulk Ni and Ni₃Al predicted by MTP, DFT, and EAM methods. The available experimental results are given for comparison. 1NN, 2NN and 3NN represent the first-, second- and third-nearest neighbors. All energies are given in eV.

_		MTP	DFT	EAM	Experiments
	$E_f(V_{\rm Ni})$	1.425	1.425	1.572	1.4±0.2[109], 1.74±0.06[110], 1.73±0.07[111]
	$E_m(V_{\rm Ni})$	1.000	1.000	1.195	1.24[112], 1.38[113]
Ni	E_f^{di-1NN}	2.835	2.799	2.890	2.92~3.10[110], 2.42[114], 2.46[115]
	$E_f^{di-2NN}(V_{\rm Ni},V_{\rm Ni})$	2.924	2.912	3.129	
	$E_f^{di-3NN}(V_{\rm Ni},V_{\rm Ni})$	2.853	2.865	3.141	
	$E_b^{di-1NN}(V_{\rm Ni},V_{\rm Ni})$	0.015	0.051	0.164	0.4±0.2[110], 0.28[114], 0.44[115]
	$E_b^{di-2NN}(V_{\rm Ni},V_{\rm Ni})$	-0.074	-0.063	0.016	
	$E_b^{di-3NN}(V_{\rm Ni},V_{\rm Ni})$	-0.003	-0.016	0.004	
	$E_f(V_{\rm Ni})$	1.588	1.643	1.773	1.6±0.2[116]
	$E_f(V_{\rm Al})$	1.710	1.787	1.544	1.6±0.2[116]
Ni ₃ Al	$E_b^{di-1NN}(V_{\rm Ni},V_{\rm Ni})$	-0.121	-0.154	-0.121	
	$E_b^{di-2NN}(V_{\rm Ni},V_{\rm Ni})$	0.068	0.107	-0.009	
	$E_b^{di-3NN}(V_{\rm Ni},V_{\rm Ni})$	0.115	0.125	-0.005	
	$E_b^{di-1NN}(V_{\rm Ni},V_{\rm Al})$	-0.048	-0.076	-0.179	
	$E_b^{di-2NN}(V_{\rm Ni},V_{\rm Al})$	-0.044	-0.043	-0.008	
	$E_b^{di-1NN}(V_{\rm Al},V_{\rm Al})$	0.031	0.053	0.000	
	$E_b^{di-2NN}(V_{\rm Al},V_{\rm Al})$	0.004	0.002	0.019	
	$E_f(Al_{Ni})$	0.957	1.081	1.436	
	$E_f(Ni_{Al})$	0.159	0.033	-0.168	
	$E_f^{1NN}(\text{Al}_{\text{Ni}},\text{Ni}_{\text{Al}})$	0.842	0.839	0.901	
	$E_f^{2NN}(\text{Al}_{\text{Ni}},\text{Ni}_{\text{Al}})$	1.098	1.080	1.237	
	$E_f^{3NN}(\text{Al}_{\text{Ni}},\text{Ni}_{\text{Al}})$	1.120	1.095	1.256	

All the formation and binding energies in the present work were calculated using a $3 \times 3 \times 3$ supercell of 108 atoms. The minimal energy path (MEP) and migration energy barrier E_m of mono-vacancy were calculated using climbing image nudge elastic band (CI-NEB) method [117].

Table III summarizes the calculated formation and binding energies of point defects and clusters in bulk Ni and Ni₃Al predicted by MTP, DFT, and EAM methods. One can observe that our developed MTP potential is capable of nicely reproducing all the DFT results, capturing both the values and the signs of the binding energies accurately. This is a remarkable outcome, since machine learning energies for defects is much more challenging than for bulk systems [65, 69, 118]. Considering the limited descriptive capacity of the EAM model, the performance of the EAM potential is acceptable and somehow unexpected. The noticeable disparities between EAM and the DFT results are evident in the description of the binding energies of vacancy clusters and formation energy of antisite defect Ni_{Al}. The EAM potential inaccurately predicts a negative formation energy of Ni_{Al}. The MTP-calculated formation energy of a vacancy in bulk Ni is 1.425 eV, in reasonable agreement with the scattered experimental data (1.4~1.74 eV) [109–111]. The mono-vacancy in bulk Ni₃Al demonstrates a slightly higher formation energy. The MTP-predicted formation energies for a Ni vacancy and an Al vacancy are 1.588 eV and 1.710 eV, respectively, suggesting the relatively easier formation of a Ni vacancy in bulk Ni₃Al. Experimental differentiation between Ni and Al vacancies is challenging, leading to an averaged formation energy of 1.6±0.2 eV [116]. It is important to note that the MTP model inherits the limitations of the underlying DFT, which demonstrates inaccuracies in predicting surface energy when using the PBE functional [119–123]. This could be linked to the precision of the predicted formation and binding energies of vacancies.

Regarding the binding energies of vacancy clusters, it is interestingly found that the divacancies in the first-nearest neighbor (1NN) in the bulk Ni exhibit a weak attractive interaction. However, as the distance between the two Ni vacancies increases, the interaction becomes repulsive. In the bulk Ni₃Al system, an opposite trend is observed for the two Ni vacancies. The two Al vacancies in the 1NN demonstrate a weak attractive interaction, whereas an Al vacancy and a Ni vacancy exhibit repulsive interactions up to the second-nearest neighbor.

Figure 4 illustrates the predicted MEPs for a vacancy migration to its 1*NN* site in both bulk Ni and bulk Ni₃Al systems using DFT, MTP, and EAM methods. It is evident that the MTP potential aligns well with the DFT predictions, whereas the EAM potential overestimates all the migration energy barriers as compared to DFT. However, the EAM-predicted migration energy barrier of a Ni vacancy is 1.195 eV, which is closer to the experimental values (see Table III). This is because

the EAM potential developed by Mishin [26] was obtained by deliberately fitting the migration energy barrier. Overall, the DFT-predicted migration energy barrier of a vacancy in Ni is greater than that in Ni_3Al , indicating a faster vacancy diffusion in Ni_3Al .



Figure 4. **a**, The MEP of a Ni vacancy diffusion in bulk Ni. **b**, The MEP of a Ni atom diffusing to the nearest Ni vacancy in bulk Ni₃Al. **c**, The MEP of a Ni atom diffusing to the nearest Al vacancy in bulk Ni₃Al. **d**, The MEP of a Al atom diffusing to the nearest Ni vacancy in bulk Ni₃Al. The DFT, MTP, and EAM predicted results are shown in gray solid lines, red dashed lines, and black solid lines, respectively.

We also computed the binding and formation energies of tri-vacancies in both Ni and Ni₃Al systems using DFT, MTP, and EAM methods. In the case of tri-vacancies in Ni, we meticulously analyzed 144 potential symmetrically nonequivalent configurations within a $3 \times 3 \times 3$ supercell. For the tri-vacancies in Ni₃Al, we extensively explored a total of 280 energetically non-degenerate configurations within a $3 \times 3 \times 3$ supercell. The results are compiled in Fig. 5. One can see that the MTP potential is able to capture the overall trend observed in the DFT results. In the case of tri-vacancies in Ni, the MTP potential correctly predicted the energetically most favor-

able tri-vacancies, which form a triangle in the {111} plane. The second energetically most stable tri-vacancies forming a line along the $\langle 111 \rangle$ direction were also accurately reproduced by MTP. The least favorable tri-vacancies were found to form a line the $\langle 001 \rangle$ direction. In the case of tri-vacancies in Ni₃Al, the developed MTP potential was also capable of accurately predicting the most and least favorable configurations. Nevertheless, given the accuracy of the MTP model, distinguishing the relative stability of the two tri-vacancies becomes highly challenging when their formation energy difference is less than 0.03 eV. This discrepancy contributes to variations in the relative stability of different tri-vacancies between the MTP and DFT predictions. It is also notable that the current MTP potential exhibits a larger error for tri-vacancies that involve Al vacancies, which is likely due to insufficient sampling of phase space encompassing the Al vacancies. This underscores the need for improving the training set in the future work. In contrast to the MTP potential, the EAM potential can only correctly predict the energetically most favorable configuration of tri-vacancies and typically shows a larger deviation with the DFT results.



Figure 5. **a**, **b**, Binding and formation energies of tri-vacancies in Ni. **c-j**. Binding and formation energies of tri-vacancies in Ni₃Al. The horizontal axis indicates the ID of the tri-vacancies configurations, which are ordered by DFT-calculated formation energies.

Furthermore, we evaluated formation energies of vacancies at the Ni-Ni₃Al interface boundary. As depicted in Fig. 6, all the three (DFT, MTP, and EAM) methods predict that the Ni vacancy at the site 1 is the most favorable. The MTP potential can effectively reproduce the DFT-derived relative stability of various vacancies, while the EAM potential significantly overestimates the formation energies. Again, within the accuracy, the current MTP struggles to correctly determine the order of stabilities for the vacancies at sites 2 and 3.



Figure 6. **a**, Formation energies of vacancies at the Ni-Ni₃Al interface boundary predicted by DFT, MTP, and EAM methods. **b**, The positions of the vacancies (red balls). The site number is indicated. The blue and gray balls represent Al and Ni atoms, respectively.

E. Ground-state configuration of vacancy clusters in Ni

Vacancy clusters in metals can be formed under various working conditions, including tensile or shear loads, irradiation damage, and so on [4, 5, 124–126], which therefore necessitates special investigation. It is widely accepted that in FCC metals, vacancy clusters prefer stacking fault tetragonal configurations and void configurations. While numerous studies explored these configurations using DFT [20, 127] or semi-empirical potentials [128–132], the stable configurations of vacancy clusters remain elusive due to the high computational cost of DFT and the limited accuracy of semi-empirical potentials. To address this issue, here we attempted at examining the

stability and ground-state configurations of vacancy clusters in nickel using our developed MTP potential.

We first accessed the accuracy of the MTP potential using $5 \times 5 \times 5$ supercell, a size affordable for DFT calculations. Figure 7 presents the formation energies and binding energies of the vacancy clusters of loop, SFT, and void types predicted by DFT and MTP methods. One can see that as compared to the DFT results, the MTP predictions generally overestimate the formation energy per vacancy, but underestimate the binding energy of the vacancy clusters. For vacancy clusters containing fewer than three vacancies, the current MTP potential fails to predict the correct relative order of binding and formation energies across the three types of vacancy clusters. We would like to note that reproducing the formation and binding energy of large defect clusters using machinelearned interatomic potentials is highly challenging due to the potential accumulation of prediction errors with increasing system sizes. Nevertheless, the overall trend of binding energy remains consistent with anticipated behavior.



Figure 7. **a**, Formation energies and **b**, binding energies of the vacancy clusters of LOOP, SFT, and VOID types predicted by DFT and MTP methods.

Then, we turned to a larger $8 \times 8 \times 8$ supercell consisting of 2048 atoms to determine the groundstate configuration of the vacancy clusters at their equilibrium volume and at 0 K. The vacancies loop with a triangular shape, void, and SFT configurations were considered. For the vacancies loop and void configurations, we conducted an exhaustive search considering all possible configurations. To identify the most stable configuration for the SFT and other possible configurations, we conducted multiple molecular dynamics annealing simulations.



Figure 8. **a**, Formation energies and **b**, binding energies of vacancy clusters of different sizes in an $8 \times 8 \times 8$ supercell of Ni predicted by MTP.

Figure 8 shows the MTP-predicted formation and binding energies as a function of the size of vacancy clusters in an $8 \times 8 \times 8$ supercell of Ni. The most stable configurations of vacancy clusters for each size are depicted in Fig. 9. For simplicity, in the following we denote the cluster of *n* vacancies as *n*v. It can be seen that the 3v cluster stabilizes the loop configuration, while 4v and 8v clusters favor the void configuration. The 5v cluster forms a 6v void configuration with an self interstitial atom (SIA) near the center. For the remaining vacancy clusters, the SFT configuration is found to be the most stable. Specifically, the ideal SFT configuration is attained for the 6v, 10v, 15v, and 21v clusters, known as magic numbers. For other vacancy clusters whose size does not align with these magic numbers, the SFT configuration is imperfect, with a neighboring vacancy.



Figure 9. Configurations of vacancies from monovacancy to vacancy cluster consisting of up to 24 vacancies in Ni. The green balls represent self interstitial atoms within the perfect lattice, the gray cubes denote the vacancy sites, and blue cubes represent the positions of atoms in the perfect lattice. Note that only the environment containing the defects is displayed.

F. Diffusion behavior of vacancy clusters in Ni

In addition to investigating the static energetics of vacancy clusters, we delved into the dynamic diffusion characteristics. In order to ensure the same vacancy concentration, we constructed an $12 \times 12 \times 12$ supercell containing twelve vacancies for each type of vacancy clusters. By doing so, we restricted ourselves to the clusters of mono-vacancy, di-vacancies, tri-vacancies, quad-vacancies, and hexa-vacancies. In other words, within each system, there are 6 clusters of di-vacancies, 4 clusters of tri-vacancies, 3 clusters of quad-vacancies, and 2 clusters of hexa-vacancies. Using our developed MTP potential we performed molecular dynamics simulations at 800 K over a duration of 2 ns and then computed the mean squared displacement (MSD) of each system. The results are shown in Fig. 10. We note that in the absence of vacancy diffusion, the atoms also underwent vibrational motion around their equilibrium positions, leading to a MSD of 0.1 Å², represented by the dashed horizontal line in Fig. 10.

We observed during the MD run that the di-vacancy and hexa-vacancy clusters are stable, keeping the cluster size from the beginning to the end. However, the tri-vacancy cluster is unstable, decomposing into a di-vacancy cluster and a nearby mono-vacancy after several migration steps. After about 1 ns, all four clusters of tri-vacancies underwent decomposition. During this process, the decomposed mono-vacancy remains unchanged, whereas the decomposed di-vacancy clusters exhibit behavior akin to regular di-vacancy clusters. The quad-vacancy clusters are also unstable. They initially broke down into a tri-vacancy cluster and a mono-vacancy after several migration steps. Finally, these tri-vacancy clusters further decomposed into a di-vacancy cluster and a mono-vacancy. Because of the decomposition process, the ultimate slopes of the MSD curves for di-vacancy, tri-vacancy, and quad-vacancy clusters are hence nearly indistinguishable. Under 800 K, the mono-vacancy is almost immobile, with only one jump observed throughout the entire trajectory. Interestingly, the hexa-vacancy clusters with the SFT structure (6v-SFT) exhibit the fastest diffusion.



Figure 10. MTP predicted mean squared displacements in the defective Ni system containing various type of vacancy clusters at a temperature of 800 K.

We conducted an in-depth analysis to understand why the 6v-SFT cluster diffuses so rapidly compared to the other five vacancy clusters under consideration. Within the 6v-SFT clusters, there are two configurations that are mutually centrosymmetric with each other. We ran another MD simulation with a shorter dump interval, containing only one 6v-SFT cluster in the system. During this simulation, we observed frequent transitions between the two configurations of the 6v-SFT cluster. The calculated transition barrier at 0 K using the CI-NEB method is approximately 0.4 eV, surprisingly low for such transitions to occur.



Figure 11. Snapshots from a MD trajectory. Note that only atoms near the SFT cluster are shown. The atoms are color-coded based on their site energies predicted by our MTP potential. The blue cubes represent the positions of atoms in the perfect lattice. The trajectory of the migrating atoms (marked by red arrows) is colorized according to simulation time. **a-c**. Snapshots of the first migration event occurring between 631.2 ps and 635.2 ps. **d-f**. Snapshots of second migration event occurring between 724.2 ps and 744.2 ps.

To uncover the migration path of the 6v-SFT cluster, we applied a low pass filter with a super-Gaussian window on the trajectory to effectively filter out thermal vibrations above 10 THz. At 631.2 ps, during the transition between the two configurations of the SFT cluster, an atom underwent a rapid and long-distance migration across the core region. It moved swiftly, covering a significant distance, and then quickly returned to its original position within a few femtoseconds. The trajectory of the migrating atom is shown in Fig. 11a-c. Notably, a hole appeared at the center of the cluster during the transition between the two symmetrically opposite configurations of the 6v-SFT, potentially aiding in the occurrence of the observed long-distance transfers.

Similarly, at 724.2 ps, a migration event similar to the one observed at 631.2 ps reoccurred. However, this time another atom took the place of the previously migrated atom, as shown in Fig. 11d-f. This caused the cluster to enter an unstable state characterized by rapid transfers of core atoms. This unstable state persisted for over 500 ps in the trajectory, corresponding to the rising section of the MSD curve in Fig. 10. Once the cluster returned to a stable 6v-SFT configuration, the rapid migrations ceased, rendering the cluster immobile once more. Following this process, the 117 atoms of the 6v-SFT cluster including the initial SFT core atoms migrated approximately 17 Å. To enhance the understanding of the migration process, we have included an additional supplemental movie.

G. Description of the energetics of planer defects by MLIP

Having assessed the accuracy of our developed MLIP in describing point defects, we now turn to evaluating its performance in the description of planer defects such as generalized stacking faults (GSF). The GSF was introduced by Vitek [133] to describe the energy variations that occur when half of a crystal undergoes shear displacement across a glide plane in specific directions. It provides valuable insights into the behavior of intrinsic stacking faults and the deformation mechanisms of materials [134, 135]. Accurate modeling of stacking faults is crucial for understanding the strength and plasticity of materials, since the intrinsic stacking fault energies and distances of these faults are critical parameters for modeling dislocations [136].

Employing our developed MTP potential, we computed the generalized stacking fault energy (GSFE) for the densely packed (111) plane in both Ni and Ni₃Al. To model the stacking faults, the shear alias method [137] was employed using a 6-layer supercell. The GSFE was calculated as

$$\gamma_{GSF} = (E_{GSF} - E_0)/A,\tag{12}$$

where E_{GSF} and E_0 are total energies of the supercell with or without the stacking fault, respectively, and A is the area of the slip plane within the supercell. For determining the GSFE on the (111) plane, a 16 × 26 grid was utilized for DFT calculations, while a dense 120 × 120 grid was applied for MTP and EAM calculations.

Figure 12 presents the GSFEs of the (111) plane of Ni and Ni₃Al predicted by DFT, MTP, and EAM methods. The values of various stacking fault energies are summarized in Table IV. Overall, the MTP predictions demonstrate better agreement with the DFT results than the EAM predictions. Given the scattered nature of both the calculated and experimental literature data, our predictions are in line with them. The MTP prediction errors are negligible in the low-energy region, but tend to increase in the high-energy region. This is due to inefficient sampling of these



energetically unfavorable configurations from unbiased molecular dynamics simulations.

Figure 12. **a**, **b**, and **c**, Contour plot of generalized stacking fault energy of the Ni (111) plane predicted by DFT, MTP, and EAM, respectively. **d**, **e**, and **f**, Contour plot of generalized stacking fault energy of the Ni₃Al (111) plane predicted by DFT, MTP, and EAM, respectively. The *x* and *y* axis denote the displacements from initial perfect lattice along the [110] and [112] directions, respectively. The color coding indicates the generalized stacking fault energy in unit of mJ/m^2 .

		DFT	MTP	EAM	Other calculations	Experiments
	$\gamma_{ m ISF}$	133	115	139	117.70	120~130[138]
Ni	$\gamma_{ m usf1}$	263	276	297	283[139],273[140],278[141]	
	$\gamma_{ m usf2}$	713	688	974	783[139]	
	$\gamma_{\rm CSF}$	210	196	238	205[142],208[16],225[143],249[15]	235±45[144]
	$\gamma_{ m APB}$	163	235	206	180[142],198[140],259[139]	175±15[144]
	γ_{SISF}	55	73	41	21[140],47[139],75[142],80[143]	6±5[144],35[145]
Ni ₃ Al	$\gamma_{ m usf1}$	1324	1291	1618	1332[140],1368[142],1421[139]	
	$\gamma_{ m usf2}$	808	788	948	778[142],791[140],830[139]	
	$\gamma_{ m usf3}$	232	248	252	227[140],254[142]	
	$\gamma_{ m usf4}$	214	299	257		
	$\gamma_{ m usf5}$	522	524	610		

Table IV. Stacking fault energies of the (111) plane of Ni and Ni₃Al predicted by DFT, MTP, and EAM methods. The specific definition of each stacking fault energy has been provided in Fig. 12. The results are compared to other calculations as well as experiments. All energies are in unit of mJ/m^2 .

Following the minimal slide path as depicted in Fig. 12, our results clearly demonstrated that in bulk Ni, a $\frac{1}{2}[1\overline{1}0]$ dislocation decomposes into two Schockley partial dislocations $\frac{1}{6}[2\overline{1}\overline{1}]$ and $\frac{1}{6}[1\overline{2}1]$ plus a intrinsic stacking fault (ISF). In the case of Ni₃Al, a [1\overline{1}0] super dislocation first decomposes into two $\frac{1}{2}[1\overline{1}0]$ dislocations plus an anti-phase boundary (APB). Then, each $\frac{1}{2}[1\overline{1}0]$ decomposes into two Schockley partial dislocations $\frac{1}{6}[2\overline{1}\overline{1}]$ and $\frac{1}{6}[1\overline{2}1]$ plus a complex stacking fault. Our results are consistent with the discussions presented in the literature [2, 145–148].

It is worth mentioning that the anti-phase boundary (APB) in Ni₃Al, as shown in Fig. 12, exhibits a slight deviation from its geometrically ideal position after structural relaxation. For this reason, we employed the minimal energy point of the APB instead of the geometrically ideal one to determine the stacking fault energies. As a result, our computed value of γ_{APB} is lower than the values reported in the literature.

H. Melting point prediction by MLIP

As a final inference test for our developed MLIP, we computed the melting points of Ni and Ni₃Al using the solid-liquid coexistence method [149]. A $10 \times 10 \times 20$ supercell consisting of 8000 atoms was employed. The solid and liquid phases were separated by the (001) interface. The calculated melting point of Ni is around 1597 K, which underestimates the experimental value of 1728 K [150, 151]. For Ni₃Al, the MTP-predicted melting point is 1630 K, in good agreement with the experimental value of 1645 K [150, 151]. We note that our training set did not deliberately incorporate melting structures. It is therefore remarkable that the MTP potential we developed accurately predicts the melting points of both Ni and Ni₃Al, showcasing a notable extrapolation capacity.

V. CONCLUSIONS

In conclusion, we have proposed an effective genetic algorithm based optimization scheme for moment tensors contractions, which significantly reduces the number of independent moment tensor components and intermediate tensor components. This results in almost an tenfold acceleration in speed and improved accuracy as compared to the traditional MTP model of Shapeev [44] for the basis sets with high level of complexity. The performance of our improved MTP model has been thoroughly assessed by predicting the energetic and dynamical properties of various point and planar defects in Ni-Al alloys. We found that our developed MTP model not only is capable of reproducing the fundamental physical properties of bulk Ni and Ni₃Al such as the lattice constants, elastic properties, energy-volume curves, phonon dispersions, and the melting points, but also can accurately predict the formation and binding energies of vacancy clusters, antisite defects, and stacking faults as well as the diffusion behavior of vacancy clusters, in general surpassing the widely used semi-empirical EAM potentials. The latter perform well for the target properties used in fitting, but tend to exhibit large errors for complex defects such as vacancy clusters and stacking faults. Our developed MTP model also enables the identification of the ground-state configuration of vacancy clusters of differing sizes, accurately predicting the optimal SFT configuration for the 6v, 10v, 15v, and 21v clusters. Interestingly, we found that the 6v cluster with the SFT configuration is very stable and diffuses fastest among the considered vacancy clusters (1v, 2v, 3v, 4v, and 6v) with the same vacancy concentration. Furthermore, our work underscores the general challenge for MLIP models to predict the correct relative stability order of the defects with comparable formation energies, which probably requires an accuracy of less than a meV per atom. While more complex basis sets can achieve this high level of accuracy, it comes at the expense of efficiency. Finally, we would like to note that the current MTP model can be systematically improved by broadening the phase space of the training set through the active learning approach, thereby laying the foundation for developing a general-purpose MLIP for Ni-Al alloys.

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