

Thermodynamics and phase stability of the AlTiCrMoW high-entropy alloy simulated using machine-learned interatomic potentials

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High-entropy alloys (HEAs) possess exceptional mechanical properties and thermal stability, yet their vast compositional space and complex atomic arrangements present significant challenges for property prediction and design. To overcome this, we have trained a machine-learned interatomic potential (MLIP) for the AlTiCrMoW HEA using the message-passing atomic cluster expansion (MACE) framework. This new MLIP is trained on a series of high-accuracy density functional theory (DFT) calculations that covers the available configuration space of the HEA. The training set includes key configurations with linear-response-informed short-range ordered configurations, which are generated using a concentration wave analysis within the coherent potential approximation (CPA) and subsequently optimized using conventional DFT calculations. Phonon dispersion and (PDBs) for the HEA are generated using the MACE model, and several physical properties are extracted for the HEA, such as the elastic constants and the bulk modulus. The fine-tuned model reproduces DFT structural and elastic benchmarks with high accuracy, at a significantly reduced computational cost, allowing for larger simulations to be performed, which can more accurately probe the configuration space and composition of the alloy. Here, we perform Monte Carlo based simulations to identify the ordering transition of equiatomic AlTiCrMoW, where we match the experimental value of 1273 K. Vacancy relaxation volumes are investigated for AlTiCrMoW, as an example of a property analysis, which would be computationally intractable via DFT.

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I. INTRODUCTION

Metallic alloys offer a controllable thermodynamic landscape in which lattice type, atomic short-range order (ASRO), and defect populations can be steered through composition to target specific deformation, transport, and environmental responses. High-entropy alloys (HEAs) exploit this principle by concentrating five or more principal elements at near-equiatomic fractions, thereby increasing the configurational entropy: $S_{\text{conf}} = -k_B \sum_i x_i \ln x_i$, where x_i denotes the atomic fraction of component i . The associated entropic contribution to the free energy, $-TS_{\text{conf}}$, lowers the free energy of the disordered solid-solution state relative to competing ordered or multiphase configurations, thereby stabilising single-phase solid solutions at high temperature. At typical processing temperatures, this entropy penalty for ordering can offset moderate enthalpies of mixing, suppress competing intermetallics and promote single-phase bcc or fcc solid

solutions [1–6]. The resulting alloys are chemically complex, concentrated solutions in which atomic local order is random, and which exhibit property combinations not readily accessible to dilute alloys, including retention of strength at elevated temperature [7], improved radiation tolerance through reduced defect mobility and recombination bias [8,9], and exceptional fracture resistance in specific systems linked to heterogeneous local bonding and slip delocalization [10,11].

Within this context, the quinary AlTiCrMoW alloy introduced by Gorr *et al.* [12] is a prototypical refractory HEA: it forms a bcc solid solution with a microhardness of about 800 HV and exhibits remarkable oxidation resistance [12,13]. The selection of chromium, molybdenum, titanium and tungsten exploits the fact that each adopts bcc above 1273 K, biasing the phase space toward a bcc matrix with long-term strength and environmental stability at and above 1273 K. Aluminium, although fcc in the pure state, is incorporated because in many refractory HEAs it stabilizes bcc and can promote B2-like short-range chemical preferences [14,15]; in the benchmark Al_xCrFeCoNi series the structure evolves toward fully bcc as the aluminium fraction increases [16–18].

Thermodynamic calculations by Gorr *et al.* place the melting point of AlTiCrMoW near 1973 K and predict bcc stability from about 1373 K up to melting [12]. Direct experimental determination of these boundaries is nontrivial because precise control of composition, thermal history, and oxidation is

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required at high temperature; hence their use of calculation of phase Diagrams (CALPHAD) [19] for the calculation of thermodynamic properties.

A lattice cluster expansion would provide an efficient description of configurational energetics on fixed crystallographic sites, and is therefore well suited to phase stability and ordering questions when atomic relaxations can be incorporated into an effective lattice Hamiltonian. The present work has a different objective. We require a continuous coordinate interatomic potential that provides energies, forces, and stresses for chemically disordered configurations, so that lattice relaxation, elastic response, phonons, and vacancy relaxation volumes can be computed within the same model. These quantities are central to the calculations reported below and cannot be obtained directly from a purely configurational cluster expansion.

While CALPHAD would be a robust option for low-order systems, its reliability degrades in near-equiatomic HEAs because the parameterized Gibbs energy functions are assessed mainly for unary, binary, and limited ternary subsystems, then extrapolated to quinary space using geometric schemes (e.g., Muggianu [20], Kohler [21], or Toop [22]) with Redlich-Kister polynomials [23]. In concentrated multicomponent alloys, the free energy is strongly influenced by higher-order cross-interactions, temperature-dependent vibrational and magnetic contributions, and chemically specific ASRO or B2-like short-range chemical preferences on the bcc lattice—effects poorly constrained by underlying databases. Predictions for phase stability, ordering transitions, and intermetallic driving forces thus vary significantly with extrapolation choices and parameters, motivating a complementary, system-specific approach that resolves local environments and configurational correlations beyond CALPHAD's mean-field treatment.

Despite the maturity of CALPHAD for binary and ternary alloys, there is no bespoke thermodynamic assessment or property prediction framework tailored to AlTiCrMoW. We are not aware of a custom parameter set, mobility database, oxidation model, or equivalent surrogate that has been validated for this exact quinary composition. In fact, to the best of our knowledge, there is no prior end-to-end modeling workflow that targets AlTiCrMoW specifically. Establishing such a workflow would deliver a quantitative map of phase stability and ordering tendencies across temperature and composition, expose ASRO and potential B2-like tendencies within the bcc field, and translate directly into processing windows and performance envelopes for high-temperature use. The lack of a vetted database for this system therefore motivates a shift from database extrapolation to simulations grounded in first-principles calculations that resolve local chemical environments and lattice distortion explicitly.

A route free of empirically determined parameters is desirable, as this naturally ensures that any developed modeling approaches are transferable across the composition space. Methods based on first-principles Density Functional Theory (DFT) calculations are an appropriate choice, as DFT is well-understood to exhibit a high degree of fidelity, accurately predicting relative phase energies, elastic moduli and defect energetics across a wide range of metallic materials. [24–26]. However, the computational cost of calculations on large

simulation cells confines the routine use of DFT to systems containing only a few hundred atoms, which limits configurational entropy, restricts chemically realistic ASRO, and precludes faithful treatment of extended defects such as dislocation cores, grain boundaries and voids. These constraints are particularly severe in concentrated HEAs, where strong coupling between local composition and lattice distortion governs thermodynamics and defect structure.

Classical interatomic potentials offer a much cheaper alternative to DFT, but come with a high cost in modeling accuracy. For metals, the standard classical potentials are Finnis-Sinclair (FS) [27], embedded atom models (EAMs) [28], and modified embedded atom models (MEAMs) [29], with the latter including bonding directionality to improve the model [29]. These are intrinsically limited by the mathematical form of the potential; they cannot, with high levels of accuracy, represent the complex quantized effects of metallic electrons. Furthermore, to the best of our knowledge, there are no available EAMs or MEAMs which are capable of modeling AlTiCrMoW, further motivating the necessity to find a model with computational cost comparable to that of classical force fields while retaining near *ab initio* accuracy. Machine-learned interatomic potentials (MLIPs) make this possible; they have been built and tested for both inorganic systems and molecular crystals [30–37] and have been used extensively to predict compositional and configurational stability of HEAs [38–40].

Among MLIPs, architectures based on message passing have demonstrated the ability to reproduce complex many-body interactions with near *ab initio* accuracy at a fraction of the computational cost. The present work employs the message-passing atomic cluster expansion (MACE) framework [41], which combines the symmetry principles of the atomic cluster expansion with the expressivity of equivariant graph neural networks. In MACE, the total energy is decomposed into atomic contributions whose local environments are represented by tensorial features constructed from spherical harmonics and radial basis functions. By coupling these features multiplicatively across message-passing layers through Clebsch-Gordan contractions, MACE systematically recovers higher-order body correlations while maintaining rotational equivariance. This formalism allows the model to represent bond-angle physics, charge redistribution, and elastic distortion effects that are inaccessible to classical EAM-style potentials.

Foundation models such as MACE-OMAT [33] are pre-trained on extensive, chemically diverse datasets and therefore provide general representations of atomic interactions. These models can be fine-tuned with a comparatively small number of *ab initio* data points to specialize them for a given alloy or thermodynamic regime [42,43]. In this work, we fine-tune the MACE-OMAT foundation model on a targeted first-principles dataset that samples the relevant configuration space of AlTiCrMoW, enabling accurate and transferable modeling of chemical disorder, lattice distortion, and thermo-mechanical response in high-entropy alloys.

With this machine-learned interatomic potential (MLIP) we address three fundamental questions that remain unresolved for AlTiCrMoW. (i) Can the temperature-dependent phase stability be probed via a Monte Carlo (MC) workflow using an MLIP, particularly the bcc stability window

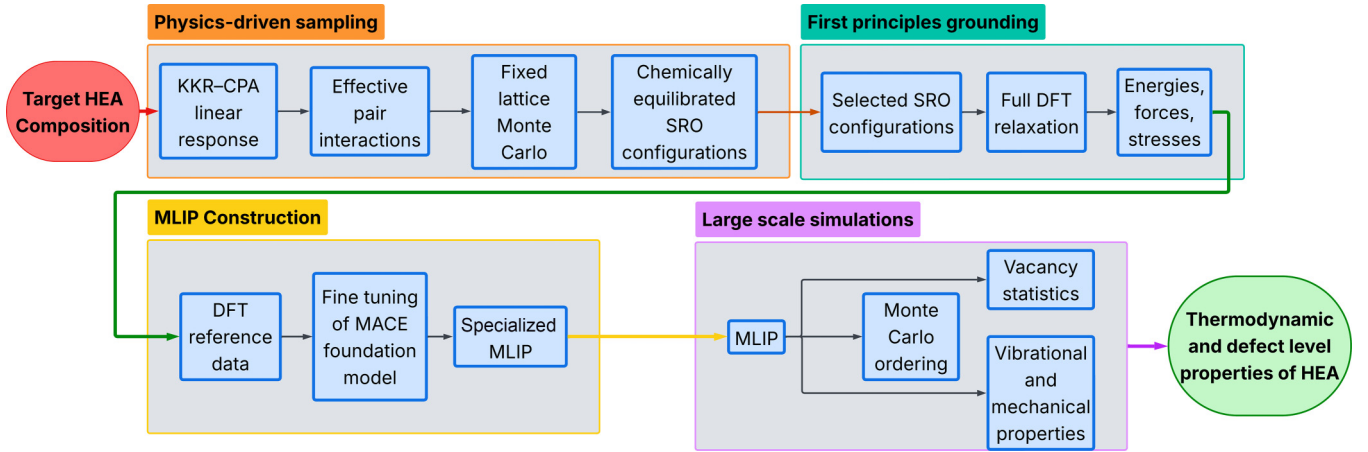


FIG. 1. Workflow used to generate ground state configurations and fine-tune the MACE MLIP.

predicted thermodynamically by Gorr *et al.*? (ii) How do short-range chemical ordering tendencies evolve with temperature, and do they correlate with the exceptional mechanical properties? (iii) Can an MLIP be used to evaluate relative stabilities of point defects in HEAs? Beyond these thermodynamic questions, we exploit the computational efficiency of the MLIP to investigate point defect properties, as may be vacancy formation energetics and relaxation volume, which govern diffusion [44] and creep at application-relevant temperatures [45] but are prohibitively expensive to sample comprehensively with DFT in chemically disordered systems.

To answer these questions, we compute Phonon Dispersion Bands (PDBs) for the equiatomic quinary alloy and perform fixed-lattice Monte Carlo simulations (to determine phase equilibria) from 1 K to 3000 K. From these calculations, we extract temperature-dependent ASRO parameters, construct a temperature-dependent ordering map, and identify the thermodynamic origins of the experimentally observed bcc stability. The approach yields atomistic predictions of ordering behavior in this alloy at application-relevant temperatures and length scales that capture realistic configurational fluctuations.

The present study brings together three elements that have not previously been combined for a five-component refractory alloy. First, Korringa-Kohn-Rostoker (KKR) coherent potential approximation (CPA) linear-response is used to guide the sampling of short-range-ordered configurations in a manner that reflects the intrinsic chemical stiffness of Al–Ti–Cr–Mo–W. Second, these configurations are relaxed at the DFT level and used to specialize a recent MACE foundation model to the narrow but highly complex configurational space of the equiatomic alloy. Third, the resulting interatomic potential is deployed to access thermodynamic and defect properties, including vacancy statistics and Monte Carlo ordering behavior, that are inaccessible to direct DFT. This integrated workflow provides a level of chemical, vibrational and configurational resolution that, to our knowledge, has not been achieved previously for this class of refractory high-entropy alloys.

The paper is structured as follows. We first detail how the training set was established and then further describe the protocols we used to train our MLIP (Sec. II). We then use our MLIP to calculate the PDBs of AlTiCrMoW and

perform fixed-lattice Monte Carlo simulations at temperatures between 1 and 3000 K to evaluate the phase stability of AlTiCrMoW and construct a temperature-dependent ordering map (Sec. IV). In Sec. III, we benchmark the resulting trained MLIP against DFT and experimental data. In Sec. V, we demonstrate the capabilities of the trained MLIP to compute vacancy energetics and study the effect the local environment has on their formation. The workflow presented in this paper is visualized in Fig. 1. Finally, we close with the conclusions and the potential future advancements in Sec. VI.

II. COMPUTATIONAL FRAMEWORK FOR TRAINING THE MLIP

The development of a reliable MLIP requires both a representative training dataset and a carefully designed training strategy. Here we summarize the workflow used to construct and refine the reference data for equiatomic AlTiCrMoW, schematized in Fig. 1. We first use KKR-CPA linear response to quantify the dominant concentration-wave instabilities and to define an on-lattice energetic model that is used solely to generate chemically representative short-range ordered configurations (Sec. II A). A subset of these configurations is then fully relaxed at the DFT level to obtain reference energies, forces, and stresses (Sec. II C), which form the ground-truth dataset employed to fine-tune the MACE-OMAT foundation model (Sec. I in SM [46]). Guided sampling is subsequently used to broaden coverage of the relevant configurational and strain space (Sec. II B), and the resulting MLIP is then deployed for the finite-temperature and defect calculations reported in subsequent Secs. III–V.

A. Generation of supercell configurations with varying atomic short-range order

To generate structures for inclusion in the training dataset for the potential for the AlTiCrMoW HEA considered in this work, we follow the protocol originally proposed by Shenoy *et al.* [47], which uses a multistage workflow to generate representative alloy supercell configurations with KKR-CPA guided ASRO. The workflow begins by performing self-consistent calculations within the KKR

formulation [48–50] of DFT, using the coherent potential approximation (CPA) [51–53] to construct an effective medium mimicking the electronic structure and consequent internal energy of the disordered solid solution.

Briefly, within the KKR formulation of DFT, the electronic structure is obtained from the single-particle Green’s function [54]

$$G(E) = \lim_{\eta \rightarrow 0} [E + i\eta - H_{\text{KS}}]^{-1}, \quad (1)$$

where H_{KS} is the usual Kohn-Sham Hamiltonian. The scattering properties of each atomic site are encoded in a t -matrix, while structural information is encoded in the free particle Green’s function, G^0 . In turn, the t -matrices for each site are concatenated to form a supermatrix for the entire system, and the scattering path operator (SPO), τ , is constructed in reciprocal space via [54]

$$\tau(E) = [(t(E))^{-1} - G^0(E)]^{-1}. \quad (2)$$

From this quantity, the full Green’s function for the system is recovered as a post-processing step [54]. For disordered alloys, the CPA [51–53] replaces the set of random atomic scatterers $\{t_\alpha(E)\}$ by an effective medium described by a coherent t -matrix $t_{\text{CPA}}(E)$. The CPA condition requires that the configurationally averaged SPO for embedding atom(s) of chemical species α into the effective medium equals that of the effective medium,

$$\sum_{\alpha} c_{\alpha} \tau_{\alpha}(E) = \tau_{\text{CPA}}(E), \quad (3)$$

with c_{α} the concentration of species α and $\tau_{\alpha}(E)$ the scattering path operator for embedding atom α into the coherent medium. Solving these equations self-consistently yields an effective electronic medium that reproduces the average electronic structure of the random HEA, and thereby provides access to the internal energy and response functions of the disordered solid solution. In this work, the HUTSEPOT package [55] was used to construct the self-consistent KKR-CPA potentials. Subsequently, the $S^{(2)}$ theory for multicomponent alloys [56–58] is used to perturbatively analyze the energetic cost of atomic-scale chemical fluctuations. These atomic-scale chemical fluctuations are described using the language of so-called “concentration waves” [59].

Effective pair interactions. Thus, from the KKR-CPA linear-response analysis, we obtain effective pair interaction (EPI) coefficients [56] that define a coarse-grained on-lattice energetic model for chemical configurations. An on-lattice configuration is specified by site occupancy variables $\{\xi_{i\alpha}\}$, where $\xi_{i\alpha} = 1$ if lattice site i is occupied by species α and $\xi_{i\alpha} = 0$ otherwise, with the constraint $\sum_{\alpha} \xi_{i\alpha} = 1$ for each site. The corresponding Bragg–Williams (pair) Hamiltonian is [60,61]

$$H(\{\xi\}) = \frac{1}{2} \sum_{i\alpha} \sum_{j\alpha'} V_{i\alpha;j\alpha'} \xi_{i\alpha} \xi_{j\alpha'}, \quad (4)$$

where $V_{i\alpha;j\alpha'}$ are the EPIs (finite-ranged and lattice translation invariant in the present approximation) obtained from the CPA perturbative formalism. Note that $H(\{\xi\})$ is used only to generate chemically representative short-range-ordered

configurations for subsequent DFT relaxation; it is not used to report final energetics.

Fixed lattice Monte Carlo. To generate chemically representative configurations consistent with the CPA-derived interactions, we then perform fixed lattice occupational Monte Carlo sampling of the Bragg–Williams Hamiltonian defined by the EPI coefficients. This step is used solely to equilibrate initially random supercells at selected temperatures under occupational exchange moves that conserve the global composition, thereby producing configurations with short-range order consistent with the CPA-derived interactions. The procedure promotes thermally activated occupational swaps on a fixed lattice geometry by sampling the canonical distribution associated with the Bragg–Williams Hamiltonian [Eq. (4)]; at each step, two lattice sites are selected, and their species exchanged; the corresponding energy change ΔH is computed from the EPI parameterized Hamiltonian, and the move is accepted with probability $\min\{1, \exp(-\beta\Delta H)\}$, where $\beta = 1/(k_{\text{B}}T)$. All atomic positions and lattice vectors are held fixed during this sampling. The implementation thus follows the standard Metropolis algorithm [62] as provided in the BRAWL package [63]; the outlined approach has already been shown to successfully capture the thermodynamics and phase stability of several Al-containing HEAs [18,64]. We emphasize that this sampling is employed only to construct a diverse set of SRO configurations for subsequent DFT relaxation and labeling; it is distinct from the MLIP-based Monte Carlo simulations described later in Sec. II G, which are used to analyze ordering trends.

The outputs of the present step are therefore (i) the temperature-dependent linear-response information contained in $S_{\mu\nu}^{(2)}(\mathbf{q}, T)$ and its dominant eigenmodes, and (ii) a set of on-lattice configurations exhibiting representative short-range order. These configurations are subsequently fully relaxed within DFT to obtain reference energies, forces, and stresses for MLIP training (Sec. II C). Thus the BW model is used solely as a low-cost generator of chemically representative SRO configurations; all reported energetics and forces are obtained from DFT or the trained MLIP.

B. Training strategy and guided sampling

This subsection clarifies how the CPA linear-response analysis is used to guide configuration generation and dataset construction. The quantities defined below serve to identify dominant ordering modes and to parametrize the effective pair interactions entering the Bragg–Williams Hamiltonian [Eq. (4)]; they are not used directly in MLIP optimization. Their role is to determine which temperatures, supercell sizes, and chemical environments are sampled when constructing the DFT-labelled training dataset.

Within the KKR-CPA linear response framework, the relevant information is contained in the second variation of the grand potential with respect to infinitesimal composition perturbations, and defines the multicomponent direct correlation matrix $S_{\mu\nu}^{(2)}(\mathbf{q}, T)$. Each lattice site i is characterized by an ensemble-averaged occupation probability $\bar{c}_{i\mu} \in [0, 1]$, satisfying $\sum_{\mu} \bar{c}_{i\mu} = 1$. The corresponding microscopic occupancy variable $\xi_{i\mu} \in \{0, 1\}$ equals unity if site i is occupied by species μ in a given microstate and zero

otherwise. Its deviation from the mean, $\delta\xi_{i\mu} = \xi_{i\mu} - \bar{c}_\mu$, represents an instantaneous fluctuation with vanishing ensemble mean, $\langle\delta\xi_{i\mu}\rangle = 0$. The coarse-grained deviation of the composition field from the homogeneous alloy, $\delta\bar{c}_{i\mu} = \langle\delta\xi_{i\mu}\rangle$, is treated within linear response as a continuous variable satisfying $\sum_\mu \delta\bar{c}_{i\mu} = 0$, and its lattice Fourier transform $\delta\bar{c}_\mu(\mathbf{q})$ describes the amplitude of a compositional wave of wave vector $\mathbf{q}$ [65].

The quadratic change in the grand potential for such a fluctuation is

$$\delta\Phi_G = \frac{1}{2} \sum_{\mathbf{q}} \delta\bar{c}_\mu^*(\mathbf{q}) [(\beta C)_{\mu\nu}^{-1} - S_{\mu\nu}^{(2)}(\mathbf{q}, T)] \delta\bar{c}_\nu(\mathbf{q}), \quad (5)$$

where $\beta = 1/(k_B T)$, and $C_{\mu\nu} = \bar{c}_\mu(\delta_{\mu\nu} - \bar{c}_\nu)$ originates from the Bragg-Williams entropy (here given in the host-dependent form). The matrix in brackets is the chemical stability matrix; its smallest eigenvalue at a given $\mathbf{q}$ defines the curvature of the free energy, and its vanishing signals a thermodynamic instability toward ordering or decomposition. In contrast, $S^{(2)}$ itself is a response function of the CPA effective medium derived from the scattering-path operator.

The short-range-order covariance matrix $\Psi_{\mu\nu}(\mathbf{q}) = \langle\delta\hat{c}_\mu(\mathbf{q})\delta\hat{c}_\nu(-\mathbf{q})\rangle$ is defined as the ensemble average of products of Fourier-transformed instantaneous fluctuations $\delta\hat{c}_\mu(\mathbf{q}) = N^{-1/2} \sum_i e^{i\mathbf{q}\cdot\mathbf{R}_i} \delta\xi_{i\mu}$. By the Wiener-Khinchin theorem [66], $\Psi_{\mu\nu}(\mathbf{q})$ is the lattice Fourier transform of the real-space covariance $\langle\delta\xi_{i\mu}\delta\xi_{j\nu}\rangle$. This expression obeys the fluctuation-dissipation relation [56]

$$\beta\Psi_{\mu\nu}(\mathbf{q}) = [(\beta C_{\mu\nu})^{-1} - S_{\mu\nu}^{(2)}(\mathbf{q}, T)]^{-1}, \quad (6)$$

so that $\Psi(\mathbf{q})$ represents the Fourier-transformed chemical correlation function. In practice, Monte Carlo sampling of the Bragg-Williams Hamiltonian (Sec. II A) generates atomic configurations whose composition covariances reproduce $\Psi(\mathbf{q})$ within the linear-response regime. These configurations are then selected for DFT relaxation and labeling.

The chemically equilibrated SRO Monte Carlo configurations generated from the KKR-CPA Hamiltonian [Eq. (4)] represent equilibrium chemical disorder at fixed temperature, but they do not account for the elastic response of the lattice to compositional inhomogeneities. In HEAs, local concentration fluctuations induce lattice distortions, as regions rich in larger atoms expand, while those with smaller atoms contract, and these strain fields have an elastic energy penalty and also reduce the chemical free energy through size-mismatch accommodation [65]. To capture this chemo-mechanical coupling, each Monte Carlo configuration is fully relaxed by DFT (Sec. II C), allowing atomic positions to adjust in response to the local chemical environment.

Within a harmonic approximation, this coupling can be understood through a quadratic free energy expansion in both concentration waves $\delta\bar{c}(\mathbf{q})$ and displacement fields $\hat{\mathbf{u}}(\mathbf{q})$. This quadratic term takes the form

$$F^{(2)}(\mathbf{q}) = \frac{1}{2} (\delta\bar{c}_\mu^*(\mathbf{q}) \quad \hat{u}_a^*(\mathbf{q})) \begin{pmatrix} J_{\text{chem},\mu\nu}(\mathbf{q}) & \Xi_{\mu b}(\mathbf{q}) \\ \Xi_{\nu a}^*(\mathbf{q}) & K_{ab}(\mathbf{q}) \end{pmatrix} \begin{pmatrix} \delta\bar{c}_\nu(\mathbf{q}) \\ \hat{u}_b(\mathbf{q}) \end{pmatrix}, \quad (7)$$

where the asterisk denotes a complex conjugate, $J_{\text{chem}}(\mathbf{q})$ is the purely compositional curvature [cf. Eq. (5)], $K(\mathbf{q})$ is the harmonic elastic stiffness, and $\Xi(\mathbf{q})$ quantifies the coupling between composition and strain. The displacement field $\hat{\mathbf{u}}$ represents the mechanically relaxed response to the composition pattern. Formally eliminating $\hat{\mathbf{u}}$ by setting $\partial F^{(2)}/\partial \hat{\mathbf{u}} = 0$ yields the Schur complement [67]

$$J_{\text{rel},\mu\nu}(\mathbf{q}) = J_{\text{chem},\mu\nu}(\mathbf{q}) - \Xi_{\mu a}(\mathbf{q}) [K^{-1}(\mathbf{q})]_{ab} \Xi_{\nu b}^*(\mathbf{q}), \quad (8)$$

which represents the free energy curvature of composition fluctuations after elastic relaxation. The subtracted term $\Xi K^{-1} \Xi^\top$ is always positive semidefinite, reflecting the fact that elastic relaxation can only lower the total energy [65]: the lattice gives way to accommodate compositional inhomogeneities, softening the effective chemical stiffness. Operationally, this is realized by performing full ionic relaxation in DFT (see Sec. II C) for each Monte Carlo generated chemical configuration: the atomic positions (and cell shape where applicable) are optimized at fixed site occupancies, thereby integrating out the displacement degrees of freedom and yielding energies and force constants on the mechanically relaxed energy surface. The DFT relaxations therefore sample the mechanically relaxed energy surface, whose quadratic response to composition fluctuations is described by $J_{\text{rel}}(\mathbf{q})$ within the harmonic approximation. Because chemical disorder breaks local symmetry, the relaxed structures exhibit heterogeneous bond distortions and internal stress fields; however, all reported DFT-relaxed configurations satisfy standard force and stress convergence criteria within the chosen tolerances. We include Eqs. (7) and (8) only to clarify that the DFT-labelled training targets correspond to the mechanically relaxed chemical energetics; these expressions are not evaluated numerically within the training workflow.

From the above perturbative analysis, it is possible to recover an approximate representation of the alloy internal energy in terms of a sum over atom-atom effective pair interactions (EPIs) [56]. An on-lattice configuration is described by a set of lattice site occupancies, $\{\xi_{i\alpha}\}$, where $\xi_{i\alpha} = 1$ if site i is occupied by an atom of chemical species α and 0 otherwise. The internal energy of a configuration is then described by the Bragg-Williams Hamiltonian [Eq. (4)].

The MLIP (see Sec. II D), built on a local equivariant architecture with cutoff $R_c = 6$ Å, is trained on energies and forces from DFT-relaxed configurations and thereby learns an effective curvature $J_\theta(\mathbf{q})$ that approximates $J_{\text{rel}}(\mathbf{q})$ for wavevectors with $\|\mathbf{q}\| \lesssim \pi/R_c$ accessible within the supercell sizes employed (see Tables I, II and III). The corresponding covariance,

$$C_\theta(\mathbf{q}, T) = k_B T [J_\theta(\mathbf{q})]^{-1}, \quad (9)$$

encodes the equilibrium concentration fluctuations over the range of accessible wave vectors. We exclude configurations near the observed change in short range ordering behavior (~ 1250 K), where the chemical stability matrix $[(\beta C)^{-1} - S^{(2)}(\mathbf{q}, T)]$ becomes nearly singular. At that point, the system explores multiple competing minima rather than small perturbations about a single basin. Configurations in the immediate vicinity of this instability are excluded from the training dataset to avoid sampling across competing minima within a single local model. This preserves well-defined

TABLE I. The list of AlTiCrMoW supercells chosen for the training and test sets of our fine-tuned MACE model. These starting structures were obtained from equilibrated MC simulations as described in Sec. II A. All configurations highlighted in blue were sheared and rattled, as described in the main text.

Supercell size	Temperature (or random)	Number of relaxed configs (training set / test set)	Number of relaxed shuffled configs (training set / test set)
$2 \times 2 \times 2$	3000 K	45/5	0
	Random	18/2	0
$3 \times 3 \times 3$	3000 K	45/5	0
	Random	9/1	0
$4 \times 4 \times 4$	300 K	9/1	9/1
	500 K	9/1	9/1
	1500 K	9/1	9/1
	Random	1/0	0
$5 \times 5 \times 5$	300 K	9/1	5/2
	500 K	9/1	0
	1500 K	9/1	7/2

force-energy relationships during MLIP training. Finally, note that Eqs. (6)–(9) are introduced to define the CPA linear response quantities used to compute the effective pair interactions entering the BW Hamiltonian; they are not used directly during MLIP optimization, but they motivate and parametrize the configuration generation step

C. Density functional theory framework

The pre-training configurations generated via the KKR-CPA analysis were optimized using plane-wave periodic DFT in order to obtain accurate total energies, atomic forces, and stresses for use as reference data in the MACE training set. DFT optimizations were performed with plane-wave-based periodic DFT as implemented in the Vienna *ab initio* simulation package (VASP) [68,69]. These calculations were used to compute the ground state energies, forces on each atom, and stresses of every configuration in our training and test sets. The MLIP can only be as accurate as the DFT results, so a consistent high-level DFT setup was employed.

All optimizations were performed in a two-step procedure. In the first step, the lattice was allowed to fully relax. In the second step, a single-point evaluation was performed on the optimized geometry, which updates the plane-wave basis set

TABLE II. The additional structures included in the test set. All such structures were obtained from single-point energy evaluations of unique configurations generated as described in Sec. II B; the number of single-point DFT evaluations corresponds to the number of distinct supercell configurations extracted from equilibrated Monte Carlo trajectories and evaluated without further ionic relaxation.

Supercell size	Temperature	Number of single- point evaluations
$3 \times 3 \times 3$	300 K	50
	500 K	50
	1500 K	50
$4 \times 4 \times 4$	3000 K	50
$5 \times 5 \times 5$	3000 K	10

to the newly relaxed lattice parameters to provide an accurate final energy. All VASP calculations were performed using the generalized gradient approximation (GGA) to the DFT exchange-correlation functional, specifically the parametrization of Perdew, Burke, and Ernzerhof (PBE) [70,71]. The projector-augmented-wave method was used for all VASP calculations with the standard supplied pseudopotentials [72].

A Monkhorst-Pack grid of $\mathbf{k}$ points was used for Brillouin zone integrations [73], with a maximum permitted $\mathbf{k}$ -point spacing of $0.10 \text{ \AA}^{-1}$ used across all sizes of simulation cell. The plane-wave energy cutoff was set to 500 eV for all supercells with 128 or less atoms and reduced to 400 eV for supercells with greater than 128 atoms, in order to reduce the computational expense without any significant compromises. The electronic self-consistent convergence criterion was set to 5×10^{-6} eV, ensuring a minimum of five electronic steps, using the blocked Davidson algorithm. This was increased to 1×10^{-7} eV for all single-point evaluations. The atomic optimization routine was initially performed for all structures using the quasi-Newton residual minimization method with direct inversion in the iterative subspace (RMM-DIIS) algorithm [69]. For certain structures, we found success in switching the optimization routine to the conjugate gradient method in cases where the RMM-DIIS algorithm was slow to converge. The convergence criterion for atomic optimization was set to $0.01 \text{ eV/\AA}$.

To extend the sampling of the potential energy surface beyond equilibrium configurations, the training set was systematically augmented with controlled off-equilibrium distortions. Three types of transformations were employed: (i) *rattling*, where all atoms were displaced randomly to achieve an average displacement of $0.1 \text{ \AA}$, broadening the coverage of high-energy optical modes; (ii) *shearing*, where optimized supercells were strained by 0.5% and 2% in the xz plane to probe low-energy acoustic deformations; and (iii) *shuffling*, in which atomic species were permuted within each supercell to generate unfavorable chemical arrangements that test the model's sensitivity to ASRO. These augmentations are introduced to broaden the local neighborhood and strain states sampled in the training data beyond the relaxed minima, thereby

TABLE III. Aluminium and titanium structures which were included in the MACE training set.

Pure metal systems						
System	Supercell size	Relaxed configs	Sheared configs	Rattled configs per shear	Total number of rattled configs	Supercells with point defects
Al - fcc	$1 \times 1 \times 1$	1	76	19	760	5
Ti - fcc	$1 \times 1 \times 1$	1	76	19	760	5
Intermetallic systems						
System	Relaxed configs			Sheared configs		
AlTi - Alpha	1			3		
AlTi - Gamma	1			3		

constraining the model's force response in the near-equilibrium basin and under moderate distortions.

Thus the reference dataset comprises three distinct classes of configurations, defined consistently throughout this work. (i) Fully relaxed configurations, obtained by ionic and cell optimization within DFT until forces and stresses satisfy the chosen convergence thresholds; these provide equilibrium energies and near-zero force labels. (ii) Off-equilibrium configurations, generated either as intermediate ionic steps along the DFT relaxation trajectory or via controlled distortions (rattling and homogeneous strain); these provide finite force and stress labels that constrain the local curvature of the potential energy surface. (iii) The corresponding unrelaxed on-lattice configurations obtained from the fixed-lattice Monte Carlo sampling described above, which are evaluated at fixed lattice positions to constrain the high-energy region of the potential energy surface and which retain a CPA derived SRO prior to structural relaxation. All energies, forces, and stresses reported in the training and test sets are taken from DFT evaluations of these configurations.

This distribution of training and test structures is summarized in Table I. Smaller $2 \times 2 \times 2$ and $3 \times 3 \times 3$ supercells were sampled at 3000 K, representing nearly random configurations with minimal ASRO, whereas the larger $4 \times 4 \times 4$ and $5 \times 5 \times 5$ cells were generated at 300, 500, and 1500 K to capture ordered states and chemically correlated fluctuations. Configurations labeled as “random” in Table I refer to structures which were generated with random assignment of atomic species to sites on the bcc lattice at the target equiatomic composition. All distorted configurations were both sheared and rattled as described above. Each configuration underwent single-point DFT evaluation to ensure that the training set contained only physically realistic energies and forces.

For each Monte Carlo derived supercell, we include multiple structural states to constrain both equilibrium energetics and near-equilibrium force response. Specifically, we include (i) the initial on-lattice configuration, (ii) the final DFT-relaxed configuration, and (iii) one intermediate configuration selected from the ionic relaxation trajectory. The intermediate structure is chosen as the ionic step for which the total force norm $\|\mathbf{F}\| = (\sum_i |\mathbf{F}_i|^2)^{1/2}$ is closest to the midpoint between its initial and final values, subject to a monotonic decrease of the total energy along the relaxation path. An auxiliary validation subset of random BCC configurations generated via the KKR-CPA methodology (Sec. II A) was reserved exclusively

for testing (Table II), ensuring that all additional data served either to extend the model's configurational coverage or to benchmark its predictive performance. For this subset, we only performed single-point evaluations and did not allow for any relaxation. This was to ensure the model can accurately predict nonzero energy regions of the PES.

Because Al and Ti crystallize in fcc and hcp structures, respectively, while Cr, Mo, and W are bcc metals, additional DFT data for pure Al, Ti, and Al-Ti intermetallic phases were incorporated to improve structural transferability (Table III). The dataset includes relaxed, sheared, rattled, and defected configurations, sampling close-packed environments absent from the bcc alloy cells. Shears were applied along the main crystallographic directions ($\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 112 \rangle$, $\langle 111 \rangle$ for Al and $\langle 10\bar{1}0 \rangle$, $\langle 11\bar{2}0 \rangle$, $\langle 11\bar{2}3 \rangle$, $\langle 12\bar{3}0 \rangle$ for Ti) with magnitudes up to 6%, each followed by random rattling to half the applied shear amplitude. Intermetallic AlTi phases in both α and γ structures were similarly strained along the six independent tensor components (xx , yy , zz , xy , xz , yz). Within the quadratic expansion $F^{(2)}$, these configurations populate portions of the chemical-elastic subspace inaccessible to bcc cells, refining both the elastic curvature K and the coupling $\mathbf{E}$. Their inclusion constrains the potential to capture variations in energy and forces under changing local topology and bonding, ensuring smooth interpolation between bcc and close-packed configurations across the relevant thermodynamic range.

All structures listed in Table III were retained in the training set, as they contain information essential for learning both the stable and unstable regions of the potential energy surface. Point defects were introduced to simulate the local strain fields arising from atomic size mismatch in the alloy, such as an aluminium atom surrounded by tungsten neighbors.

With this construction, the training set spans relaxed, strained, and chemically perturbed regions of configuration space, thereby providing the MACE potential with sufficient information to learn the relevant physics of AlTiCrMoW across low- and high-energy regimes. The training and test sets comprises a total of 2962 and 757 structures, respectively. This DFT-labelled dataset defines the ground-truth energy, force, and stress targets used to fine-tune the MACE foundation model described next.

D. Fine-tuning the MACE foundation model

The MLIP employed in this work is based on the MACE framework [41]. MACE approximates the total potential

energy as a sum over strictly local atomic contributions,

$$E_{\text{tot}} = \sum_i E_i(\{\mathbf{r}_j, \alpha_j\}_{j \in \mathcal{N}_i}), \quad (10)$$

where $\mathbf{r}_j$ and α_j are the positions and chemical species of atoms in the neighborhood $\mathcal{N}_i$ of atom i . Each atomic contribution E_i is modeled from the local environment around each atom, which in turn is produced by a sequence of message-passing layers that iteratively build symmetry-aware, many-body descriptors of the local environment.

To construct the descriptor or the “feature,” at the first stage, a permutation-invariant, direction-preserving two-body “**A** features” for each neighbor j of central atom i is formed. These features consist of radial basis functions $R_n(r_{ij})$ multiplied by spherical harmonics $Y_{\ell m}(\hat{\mathbf{r}}_{ij})$; the radial functions encode distance dependence while the spherical harmonics encode angular direction. Higher-order “**B** features” are obtained by taking tensor products of **A** features belonging to different neighbors and symmetrising them with generalized Clebsch-Gordan coefficients. The symmetrization enforces $O(3)$ equivariance so that the features transform as proper spherical tensors under rotations. Physically, a **B** feature of correlation order ν represents a joint descriptor of ν neighbors and thus carries information about $(\nu + 1)$ -body couplings (the central atom plus ν neighbors): bond angles, torsional correlations, and other local many-body curvatures that determine the atomic Hessian inside the cutoff sphere. Under mild assumptions on the underlying two-body basis, the **A** features and the derived higher-order **B** features constitute a complete basis for representing many-body interactions [74], while still being computable at a cost comparable to pairwise interactions.

At message-passing layer t , messages to atom i are formed by element- and symmetry-dependent linear combinations of these **B** features [41],

$$m_{i,kLM}^{(t)} = \sum_{\nu} \sum_{\eta_{\nu}} W_{z_i k L, \eta_{\nu}}^{(t)} \mathbf{B}_{i, \eta_{\nu} k L M}^{(t)}, \quad (11)$$

where z_i is the chemical species of the receiving atom, k indexes feature channels, L indexes the angular-momentum (symmetry) channel, $W^{(t)}$ are learnable weights, ν labels the symmetry-adapted basis construction channels, and η_{ν} is a composite index enumerating the linearly independent many-body **B** features within channel ν . The message $m_i^{(t)}$ is not a secondary computational object, but the core learned representation of the local potential-energy landscape. Each message encodes the many-body geometric information of the neighborhood of atom i in a symmetry-adapted form, and the final atomic energy is obtained by applying a readout to these message-derived features. Thus, learning how to construct the messages is effectively equivalent to learning how local atomic arrangements map to energy. Because the messages incorporate higher-order correlations within the cutoff and respect the full $O(3)$ and permutation symmetries, the model acquires an expressive and physically consistent description of the interatomic interactions. In this sense, “learning the message” is how MACE learns the underlying potential energy surface (PES). For further details, the reader is referred to Ref. [41].

As mentioned above, because each **B** feature of order ν depends jointly on ν neighbors, the resulting message encodes explicit $(\nu + 1)$ -body interactions in a single layer. It is important to distinguish this *body order* (the number of neighbors coupled jointly in a single feature) from the *receptive-field depth* (the spatial radius over which information can propagate by sequential message passing). In conventional message passing neural networks (MPNNs), the only way to assemble effective high-order or long-range couplings is by stacking many layers: after t layers a node can be influenced by atoms up to roughly t graph hops away, so the spatial reach grows with depth. MACE decouples these concepts by constructing high body-order features *within one layer*—via tensor products of neighbors inside the fixed cutoff—so that rich local many-body expressivity does not require deep stacking. Consequently, MACE can represent complex local curvature (accurate local Hessians and anharmonic couplings) efficiently while keeping the number of sequential layers small. This decoupling reduces training sample complexity and avoids the propagation-depth bottleneck common to deep GNNs.

The decoupling, however, has a clear physical limit: body-order inside a finite cutoff cannot substitute for spatial reach. Collective phenomena with intrinsic length scales larger than the structural cutoff—long-wavelength acoustic modes, extended elastic strain fields around defects, and electrostatic interactions—require either (i) an increased cutoff, (ii) additional message-passing layers to propagate information from distant atoms, or (iii) explicit long-range terms (e.g., Coulombic or continuum-elastic corrections). Finally, tensor products and Clebsch-Gordan contractions enable efficient, GPU-friendly algorithms [41]. The balance between accuracy and computational efficiency is set by truncating the radial and angular basis functions and by restricting the maximum correlation order ν , which together determine the resolution and body-order richness of the model. Forces and stresses are obtained analytically by automatic differentiation of the total energy with respect to atomic positions and cell strain, ensuring energy conservation and enabling direct, stable use in molecular-dynamics and free-energy simulations.

To model the PES of the target alloys, we fine-tuned the MACE-OMAT foundation model. To balance accuracy with computational efficiency of both training and inference tasks, we fine-tuned the small-omat-0 model. This choice is intentionally conservative: the model is a compact member of the MACE-OMAT family, with scalar features, two message passing layers, and a moderate cutoff, chosen to balance accuracy with the cost of the Monte Carlo and vacancy calculations reported below. In this setup, atomic interactions were truncated at a cutoff of 6 Å, with two message passing layers allowing information to propagate through neighbors of neighbors within the cutoff graph, which encompasses the neighbor list to physically relevant shells and results in a relatively fast model. Angular features were represented using spherical harmonics up to $l = 3$, and correlations of order three were included at each layer to capture many-body interactions that are essential for describing metallic interactions. The embedding dimension was set to 128 channels, and the network contained two message-passing layers, which allowed each atom to gather information not only from

its direct neighbors but also from the neighbors of those neighbors within the cutoff distance. The maximum equivariance order (max_L) equals zero, which makes all features rotation-invariant. While higher max_L values can in principle improve accuracy, this choice reduces computational cost and ensures stable training, and we show in the results section and Fig. S1 in SM [46] that $\text{max_L}=0$ provides sufficient accuracy for the present application.

The fine-tuned model contains 8 222 254 trainable parameters that collectively define the many-body, $\text{SO}(3)$ -equivariant representation of the PES. These learnable weights parametrize the geometric message-passing operations—including radial embeddings (ten Bessel basis functions with Agnesi distance transform), tensor-product convolutions, and readout layers—rather than a fixed analytic functional form. This provides a transparent measure of model capacity and allows direct comparison with the EAM potential (250 spline coefficients), highlighting the fundamental difference in how the two approaches represent interatomic interactions.

Training was carried out for a total of 75 epochs, divided into two stages. In the first stage, which covered 75% of the epochs, the loss function was weighted with energy weight = 1.0 and forces weight = 100.0, putting greater emphasis on forces. This stage prioritizes learning the curvature of the potential energy surface and ensures that atomic forces, which dominate the dynamics, are captured with low force error. In the second stage, covering the remaining 25% of epochs, the weights were shifted to energy weight = 1000.0 and forces weight = 100.0. This adjustment emphasizes accurate total energies, helping the model better capture the relative stability of local minima while retaining the force accuracy learned in stage 1. 10% of the training data was used for validation purposes, and the rest for training.

Energies were referenced using isolated-atom energies, defined as single-point calculations of individual atoms in sufficiently large simulation cells to avoid self-interactions. This reference choice removes the arbitrary constant offset inherent in total-energy models and aligns the training targets with the convention used for computing formation energies. Importantly, this is a matter of convention only: the choice of energy reference does not affect energy differences, forces, or other physical observables learned by the model. Since “small-omat-0” employs the ScaleShiftMACE model, the absolute energies are in any case scaled and shifted during training, but the use of free atom reference states still ensures that relative formation energies are consistent and physically meaningful. Altogether, the combination of a compact model architecture, a staged training strategy, domain-relevant reference states, and targeted fine-tuning allowed us to obtain an MLIP that is accurate enough for multiphase, multicomponent systems, while remaining computationally efficient and stable in MC simulations.

E. Parametrization of EAM potentials for HEAs

An EAM [29] was fitted using the same training set as MACE in order to provide a baseline, lower-level comparison within our bulk properties results. No additional elemental or binary data were used for the EAM fit. The fitting was

carried out with the POTFIT program [75–77], in which all pairwise interactions between the five constituent elements of the AlTiCrMoW alloy were parametrized. The EAM fit is used here as a transparent low-order baseline rather than as an exhaustive search for the best possible classical potential for AlTiCrMoW. Its role is to quantify the limitations of a radial scalar density representation when fitted to the same reference data used for MACE.

In its standard form, the EAM expresses the total energy as

$$E_{\text{tot}} = \sum_i F_{\alpha(i)}(\rho_i) + \frac{1}{2} \sum_{i \neq j} \phi_{\alpha(i)\beta(j)}(r_{ij}), \quad (12)$$

where $\alpha(i)$ denotes the species occupying site i , $\phi_{\alpha\beta}(r)$ is the pairwise interaction between atom types α and β , and the host electron density at site i is

$$\rho_i = \sum_{j \neq i} f_{\beta(j)}(r_{ij}). \quad (13)$$

The embedding function $F_{\alpha}(\rho)$ is intended to encode the energy cost or gain of placing an atom of type α into the background electron density ρ_i generated by its neighbors. For a five-component alloy such as AlTiCrMoW, this formalism requires five embedding functions $F_{\alpha}(\rho)$, five density kernels $f_{\alpha}(r)$, and fifteen pair potentials $\phi_{\alpha\beta}(r)$.

Implicit in this construction are two crucial approximations. First, the local electron density ρ_i is expressed as a linear superposition of fixed atomic kernels $f_{\beta}(r)$, so that each neighbor contributes an invariant density independent of its own environment. Secondly, the embedding function $F_{\alpha}(\rho_i)$ depends only on the scalar magnitude of ρ_i , without reference to the chemical identity or angular arrangement of neighbors. These approximations perform well for elemental metals and simple binaries, where bonding environments are relatively uniform, but become increasingly restrictive in multicomponent systems.

In HEAs, chemically distinct environments can yield identical total densities and are therefore assigned the same embedding energy,

$$F_{\alpha}\left(\sum_j f_{\beta}(r_{ij})\right) = F_{\alpha}\left(\sum_j f_{\beta'}(r_{ij})\right) \quad \text{whenever} \quad \sum_j f_{\beta}(r_{ij}) = \sum_j f_{\beta'}(r_{ij}), \quad (14)$$

even when $\{\beta\} \neq \{\beta'\}$. This degeneracy neglects variations in d -band filling, charge transfer, and local screening that distinguish, for instance, Cr-rich from Mo-rich neighborhoods. The frozen kernel approximation also prevents the electronic density from adjusting to local chemical potentials or strain, leading to misrepresentation of size mismatch relaxation and charge redistribution effects observed in HEAs.

Because the EAM formalism lacks explicit angular dependence, it cannot distinguish between different local coordination geometries: both the pair term $\phi_{\alpha\beta}(r)$ and the embedding term $F(\rho)$ are purely radial, so that directional bonding from d -orbital hybridization or sp mixing cannot be captured. The MEAM [29] introduces partial angular densities but retains the same fixed-kernel form and thus omits

self-consistent screening. Consequently, chemical and elastic anisotropies are only approximately described.

The challenge is one of transferability—a fundamental issue for both EAM and MLIP approaches. Each element is represented by a single set of functions F_α , f_α , and $\phi_{\alpha\beta}(r)$, yet the combinatorial diversity of local configurations in a five-component alloy far exceeds what can be parametrized from finite data. The following schematic expressions are included only to clarify the formal limitations of the EAM baseline; they are not used in the fitting procedure. Formally, the missing cross-species curvature terms

$$\epsilon \sim \left| \frac{\partial^2 E}{\partial n_\alpha \partial n_\beta} \right|_{\text{DFT}} - \left| \frac{\partial^2 E}{\partial n_\alpha \partial n_\beta} \right|_{\text{EAM}} \quad (15)$$

quantify the residual error associated with unrepresented chemical couplings, which directly affect segregation, ASRO, and vibrational properties through the force-constant tensor (see Sec. II F). While MLIPs can in principle capture these cross-species interactions more flexibly through their many-body descriptors, they face analogous transferability constraints: accuracy depends critically on whether the training data adequately samples the relevant chemical compositions, local environments, and strain states encountered during simulation. The key practical difference lies in the degree of flexibility: MLIPs offer substantially more expressive capacity (8.2 million parameters versus 250 spline coefficients in our implementation) to represent complex multicomponent energy landscapes, but this advantage materializes only when training data comprehensively spans the target configuration space [78,79].

$$\Phi_{\alpha\beta}^{\mu\nu}(0, l) = \frac{\partial^2 E}{\partial u_{\alpha}^{\mu}(0) \partial u_{\beta}^{\nu}(l)}. \quad (16)$$

Thus, while the EAM remains computationally efficient and adequate for homogeneous metals, its simplifying assumptions make accurate parametrization of chemically complex HEAs inherently difficult. As we show in Sec. III, EAM results are thus of much lower accuracy than MLIPs such as MACE, which incorporate angular descriptors and non-linear many-body correlations, overcome these structural limitations and provide a systematic route to transferable accuracy across diverse local environments.

The full EAM functional form was fitted simultaneously, including all three constituent functions: the pair potential $\Phi(r)$, atomic electron density $\rho(r)$, and embedding function $F(\rho)$. Each function was represented using cubic splines with 10 knot points uniformly distributed over the range 0.0–6.0 Å for the pair potentials and density functions. The embedding functions were similarly discretized with 10 knot points spanning the range of electron densities sampled in the training configurations. Smooth cutoff conditions were enforced at the outer boundaries of all radial functions. All spline coefficients were treated as free parameters during optimization. This parametrization yields a total model capacity of 250 independent parameters: 150 for the 15 unique pair potentials, 50 for the 5 atomic density functions, and 50 for the 5 embedding functions.

F. Phonon calculations in disordered AlTiCrMoW

Phonon dispersions and densities of states were computed within the harmonic approximation using PHONOPY via the `phonolammps` interface [80,81]. For each chemically disordered configuration, the atomic positions and cell degrees of freedom were relaxed in LAMMPS with the base and fine-tuned MACE models at zero pressure. Harmonic force constants were then obtained by finite displacements of magnitude $\Delta u = 0.01$ Å applied to symmetry inequivalent atoms, with the induced forces evaluated by the corresponding MLIP. The acoustic sum rule was enforced in the post-processing stage within PHONOPY.

To limit the compositional bias arising because 54 is not divisible by five, we used five near equiatomic $3 \times 3 \times 3$ supercells in which each element appears ten times in one configuration and eleven times in the other four, so that the arithmetic average over the five configurations is exactly equiatomic. For each configuration k we computed $\omega_n^{(k)}(\mathbf{q})$ and $g^{(k)}(\omega)$, and report the mean dispersions and densities of states together with configuration-induced variability. Full details of the relaxation protocol, force constant construction, dynamical matrix formalism, and ensemble averaging procedure are provided in Ref. [46].

G. Monte Carlo sampling of configurational order

To probe the phase stability of the AlTiCrMoW HEA with the MACE potential, we performed Metropolis Monte Carlo simulations using Kawasaki dynamics [82], which conserve the global composition by restricting moves to atom-atom swaps. The simulations were initiated from random equiatomic configurations generated with ATOMSK [83].

At each Monte Carlo step, two lattice sites i and j occupied by species p and q respectively were selected. A trial move was proposed in which the two species were exchanged, producing a candidate configuration C' from the current configuration C . The energy difference was computed as

$$\Delta U = U(C') - U(C), \quad (17)$$

where U denotes the configurational internal energy obtained from the MACE potential at fixed cell volume and shape. The Monte Carlo simulations are intentionally performed on a fixed lattice, as the Hamiltonians describe configurational energies on an ideal bcc lattice, with local atomic relaxations introduced only at the subsequent DFT and MLIP relaxation stages to preserve a well-defined configurational ensemble.

The Metropolis acceptance rule enforces detailed balance in the canonical ensemble:

$$\mathcal{A}(C \rightarrow C') = \begin{cases} 1, & \Delta U \leq 0, \\ \exp(-\beta \Delta U), & \Delta U > 0, \end{cases} \quad \beta = \frac{1}{k_B T}. \quad (18)$$

Accepted moves update the configuration; rejected moves leave the system unchanged. Repeated sampling generates a Markov chain of configurations distributed according to the Boltzmann probability

$$\pi(C) = \frac{e^{-\beta U(C)}}{Z}, \quad Z = \sum_C e^{-\beta U(C)}. \quad (19)$$

Each simulation was run until equilibrium was achieved, assessed by convergence of energy and ASRO parameters. We analysed chemical ordering through the Warren-Cowley ASRO parameters [84,85],

$$\alpha_n^{pq} = 1 - \frac{P_n^{pq}}{c_q}, \quad (20)$$

where n labels the neighbor shell, c_q is the global concentration of species q , and P_n^{pq} is the conditional probability that a neighbor of type q occupies a site in the n th shell around a reference atom of type p . These probabilities were evaluated from the ensemble of Monte Carlo configurations as

$$P_n^{pq} = \frac{1}{n_{\text{pair}} z_n} \sum_{i \in p} \sum_{j \in \mathcal{N}_n(i)} \delta_{\sigma_j, q}, \quad (21)$$

with n_{pair} the number of atoms of type p , z_n the coordination number of shell n , $\mathcal{N}_n(i)$ the set of neighbors of site i at shell n , and $\delta_{\sigma_j, q}$ the Kronecker delta selecting species q . By construction, $\alpha_n^{pq} = 0$ corresponds to a perfectly random alloy, $\alpha_n^{pq} > 0$ indicates avoidance of p - q pairs on shell n , and $\alpha_n^{pq} < 0$ signals preference for such pairs.

This Monte Carlo procedure is therefore not merely a sampling tool but a means of testing whether the MACE potential encodes the correct balance of chemical interactions to reproduce equilibrium alloy thermodynamics. By driving the system through an ensemble of swap dynamics, we obtain ASRO parameters that quantify the extent to which species associate or avoid each other at finite temperature. These quantities are direct indicators of ordering tendencies and incipient ordering transitions, which are inaccessible to single-configuration relaxations. In this way, the MC framework links the microscopic energetics learned by MACE to macroscopic thermodynamic predictions, providing a stringent test of its ability to capture phase stability in chemically complex HEAs.

III. VALIDATION AND COMPARATIVE ASSESSMENT OF MACHINE-LEARNED AND EMBEDDED-ATOM POTENTIALS

In order to assess the fidelity of the machine-learned potential, we benchmark its predictions against DFT reference data for key bulk properties of AlTiCrMoW. We begin by comparing lattice parameters, cell volumes, and elastic constants, which provide a stringent test of the model's ability to reproduce equilibrium structural and mechanical response. The machine-learned results are then contrasted with a classical EAM baseline to highlight the limitations of conventional analytic forms in chemically complex alloys. Finally, we extend the comparison to lattice dynamics, using phonon dispersions and densities of states as a transferability test that probes both long-wavelength elasticity and local force-constant environments.

The principal benchmarks demonstrate that the fine-tuned model captures the relevant equilibrium and elastic response of AlTiCrMoW. The predicted lattice parameter agrees with both DFT and the experimental value of Gorr *et al.* within the reported uncertainty, the bulk modulus is reproduced within the DFT error bars, and the energy-volume curves

agree with DFT to within the meV per atom scale over the volume range considered. These tests are not intended as isolated bulk checks alone; they establish that the curvature of the potential energy surface is sufficiently accurate for the subsequent phonon, Monte Carlo, and vacancy calculations. For example, the fine-tuned model gives a bulk modulus of 185.5 ± 3.5 GPa compared with the DFT value 188.2 ± 2.1 GPa, whereas the base model overestimates it by about 20 GPa.

A. MACE optimizations for computation of bulk properties

All optimizations performed using the MACE model were carried by a consistent annealing-quench-minimization protocol. Each random supercell was first equilibrated using classical molecular dynamics with the MACE potential in LAMMPS. The relaxation protocol consisted of (i) an NPT anneal at 500 K under zero external pressure using anisotropic pressure control for 15 ps, allowing both atomic positions and cell parameters to adjust, (ii) an NPT quenching where the system was gradually cooled from 500 to 0.01 K over 10 ps, (iii) a conjugate-gradient energy minimization until both the maximum residual force and total energy gradient were converged below stringent thresholds ($< 10^{-6}$ in reduced units).

B. Bulk properties

The fine-tuned OMAT model reproduces DFT lattice parameters and cell volumes for AlTiCrMoW with higher accuracy than the base model, closely matching reference values across 300 optimized $5 \times 5 \times 5$ (250-atom) random configurations, as shown in Fig. 2. These configurations constitute an independent benchmark set, generated separately from the training and test datasets by random assignment of atomic species to sites on the bcc lattice at the target composition. These configurations were relaxed using the respective interatomic potentials to probe model behavior over a broad range of unseen local environments. DFT reference values are shown only where explicitly indicated, namely for the subset of nondefective DFT configurations (perfect, defect-free configurations) included in the training and test datasets. Compared with DFT, the base OMAT model slightly underestimates the lattice constant (-0.15 Å), while the fine-tuned model slightly overestimates it ($+0.10$ Å), but in both cases the deviations are small. Volume statistics confirm the superiority of the fine-tuned model, whose mean is within $+0.30$ Å³ of DFT and whose variance closely overlaps with the DFT distribution, compared with the broader fluctuations predicted by the base model.

The predicted lattice parameters show excellent agreement with experimental measurements. Gorr *et al.* [12] report a room-temperature (298 K) lattice parameter of 3.107 ± 0.013 Å from x-ray diffraction across multiple reflections. Our 0 K DFT calculations predict 3.114 Å, while the fine-tuned OMAT model yields 3.124 Å, agreeing within experimental and computational error margins.

Elastic properties reinforce this trend. Figure 3 and Table S2 in SM [46], compare bulk moduli and full elastic tensors obtained from DFT and both OMAT models. The fine-tuned

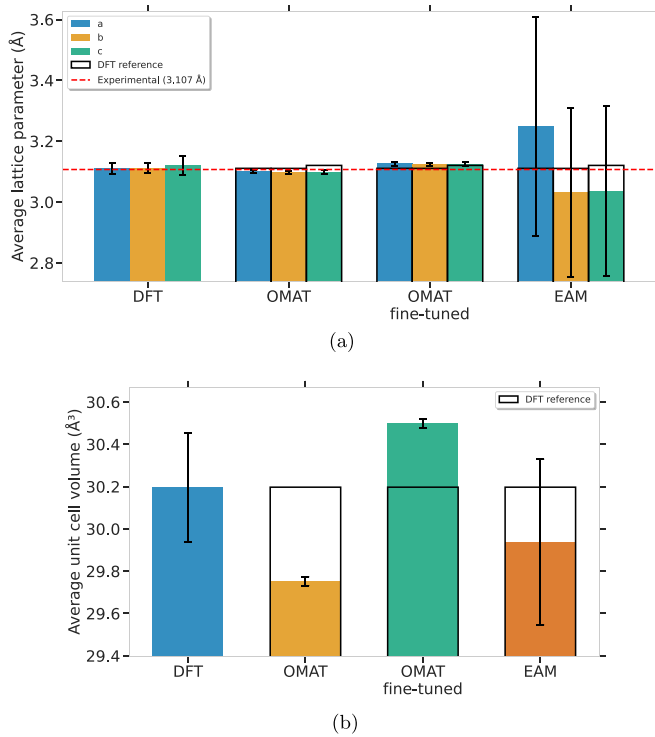


FIG. 2. (a) Average lattice parameters and (b) average cell volumes of AlTiCrMoW from DFT, the OMAT base model, and the fine-tuned OMAT model. Error bars indicate standard deviations across (i) all nondefective DFT structures included in our training set and (ii) 300 MACE optimized $5 \times 5 \times 5$ (250-atom) random configurations via both models. Values reduced to a conventional bcc unit cell.

model reproduces the DFT bulk modulus within error bars, while the base model consistently overshoots by 15–20 GPa. For the off-diagonal elastic constants, the fine-tuned model shows closer agreement than the base model, although some diagonal terms remain slightly offset due to residual lattice parameter discrepancies. Overall, fine-tuning systematically improves the overlap of the predicted elastic tensor with the DFT reference, demonstrating transferability from equilibrium cell statistics to anisotropic stress-strain response.

The equation of state analysis in Fig. 4 further substantiates this conclusion. The DFT results average three unique $3 \times 3 \times 3$ (54-atom) configurations, whilst both MACE models average ten unique $5 \times 5 \times 5$ (250-atom) configurations each. Here, unique refers to distinct chemical realizations (different random site occupations) at fixed supercell size. Only a small number of such realizations were used for the DFT equation-of-state comparison because each DFT volume scan requires multiple single-point calculations per configuration. MLIPs allow for larger samples at negligible additional cost, so more realizations are considered in this case. Ensemble-averaged energy-volume curves show that both models reproduce the DFT equation of state within 0.5 meV/atom across a wide range of volumes, with the Birch-Murnaghan [86] bulk modulus from the fine-tuned model fully overlapping with the DFT range. The base model, by contrast, deviates by up to 18 GPa. These results confirm that fine-tuning not only improves

average structural descriptors but also corrects the curvature of the energy surface, which governs elastic and thermodynamic response.

By contrast, the EAM baseline fit produces lattice constants that deviate systematically from DFT, overestimating a while underestimating b and c , leading to unphysical anisotropy in average cell shape. Although the mean cell volume partially overlaps with DFT, the variance is far larger, and the anisotropy is exaggerated. These errors reflect the intrinsic limitations of the EAM framework discussed in Sec. II E: its scalar density descriptor and radial pair interactions suppress chemical specificity and fail to penalize anisotropic distortions. As a result, the EAM permits large shape fluctuations with minimal energetic cost, whereas both DFT and the fine-tuned OMAT model predict a stiffer, more isotropic response. This comparison underscores why the EAM, while computationally cheap, cannot serve as a predictive tool for chemically complex alloys, and why machine-learned models with explicit angular descriptors are required to capture bulk structural behavior in AlTiCrMoW.

Energy convergence with supercell size shows that cells of at least 250 atoms ($5 \times 5 \times 5$) provide a stable representation of the average energetics of AlTiCrMoW, whereas smaller 54- and 128-atom cells yield only approximate agreement and 16-atom cells exhibit substantial variability. This behavior arises primarily from compositional discreteness in small supercells, where exact equiatomicity cannot be maintained, and individual configurations deviate significantly from the target composition. As the supercell size increases, both composition bias and finite-size fluctuations are reduced, leading to more consistent average energies. Both OMAT models reproduce the DFT mean energies within 50 meV/atom across all cell sizes, with the fine-tuned model consistently closer to the DFT reference. These comparisons, illustrated in Fig. 5, confirm that fine-tuning improves the predictive fidelity of the base foundation model.

C. Phonon dispersion diagrams

Phonon dispersion relations and densities of states provide a sensitive diagnostic of interatomic potentials, as they probe both the long-wavelength elastic response and the local force-constant environment in chemically disordered alloys. In the present work, phonon calculations are not intended as fully converged vibrational spectra for thermodynamic analysis, but rather as a validation of the local curvature of the potential energy surface, in particular the acoustic branches that determine elastic constants, sound velocities, and anisotropy.

Direct DFT phonon calculations for AlTiCrMoW are computationally prohibitive due to the large supercells required to represent chemical disorder. We therefore employ the MACE models to compute representative phonon dispersions as an indirect validation of their physical fidelity. The simulated PDBs for the base and fine-tuned OMAT models are shown in Fig. 6. These calculations were performed using a limited set of chemically symmetrized, near-equiatomic supercells, which is sufficient to obtain well-converged long-wavelength acoustic behavior while avoiding systematic composition bias.

As shown in Fig. 6, both models reproduce the expected qualitative features of the vibrational spectrum, with acoustic

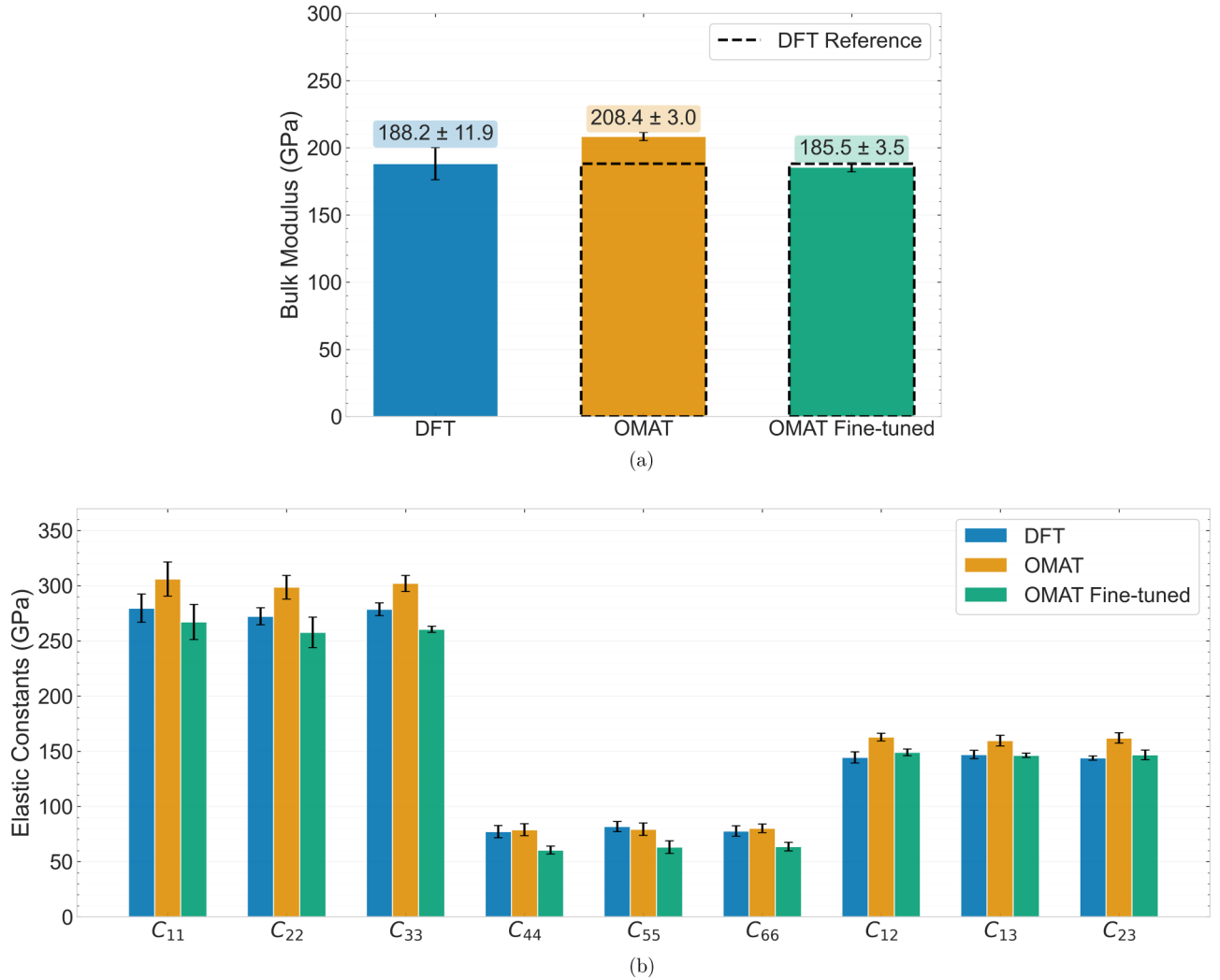


FIG. 3. (a) Bulk modulus and (b) elastic constants of AlTiCrMoW from DFT, the OMAT base model, and the fine-tuned OMAT model. Error bars denote standard deviations across $3 \times 3 \times 3$ (54-atom) supercells.

branches emerging from the Γ point and a broad manifold of higher-frequency optical modes spanning approximately 2–11 THz. The fine-tuned model exhibits slightly smoother and more coherent acoustic dispersions, while the optical branches are somewhat more clearly delineated, though of comparable overall bandwidth to those of the foundation model. The root mean square errors (RMSEs) between the two PDBs are 0.20 THz for the acoustic branches and 0.51 THz for the optical manifold. These differences indicate a modest improvement in the consistency of long-range force constants and local environment sensitivity upon fine-tuning, while the overall spectral characteristics remain similar between the two models.

We emphasize that larger deviations in the optical branches are expected in a chemically complex high-entropy alloy, where strong disorder leads to mode broadening and localization at short wavelengths. Such high-frequency modes are intrinsically short-lived and are neither well resolved experimentally nor central to classical molecular dynamics, where they are effectively filtered by the integration time step. Increasing the number of configurations or the supercell size

would primarily refine these short-wavelength optical features, while leaving the long-wavelength acoustic branches that govern elastic and transport-relevant properties essentially unchanged.

Experimental phonon dispersion data are not currently available for AlTiCrMoW, precluding direct validation. Nevertheless, given the demonstrated accuracy of the fine-tuned model in reproducing bulk structural and elastic properties, we expect the acoustic portion of the dispersion in Fig. 6 to be physically robust, while recognizing that further experimental work would be required to assess the detailed structure of the optical modes.

The phonon densities of states obtained from both models peak in similar frequency ranges. The fine-tuned model exhibits a slightly broadened envelope and an enhanced low-frequency tail, consistent with a redistribution of vibrational states associated with chemical heterogeneity and quasilo-calized modes. While these features are qualitative, they are indicative of the disorder-induced vibrational landscape characteristic of high-entropy alloys and are relevant for understanding trends in lattice thermal transport.

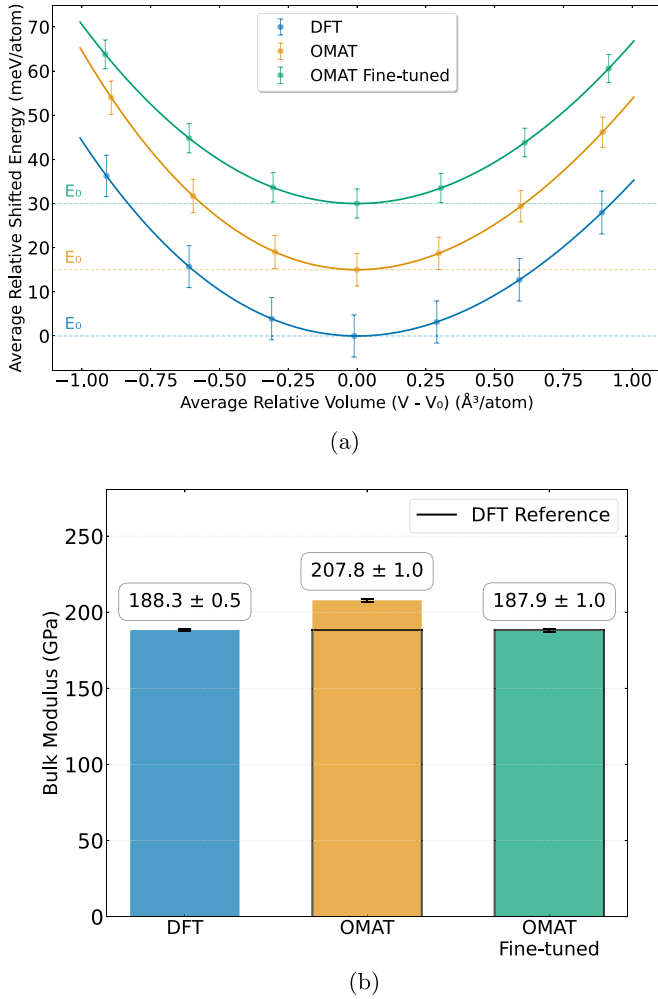


FIG. 4. (a) Energy-volume equation of state. Each E_0 value represents the zero-energy level for each respective level of theory/model. (b) Bulk moduli from Birch-Murnaghan fits for AlTiCrMoW, comparing DFT, the OMAT base model, and the fine-tuned OMAT model.

IV. CONFIGURATIONAL ORDER AND ORDERING TRANSITION VIA MACE

Having established the fidelity of the fine-tuned OMAT model against DFT for equilibrium and elastic properties, we now employ it to probe finite-temperature configurational order in AlTiCrMoW. In particular, we use Monte Carlo simulations (Sec. II G) to quantify ASRO and observed changes in short-range ordering behavior, thereby assessing whether the model predicts ordering changes consistent with thermodynamic expectations.

The lattice for these simulations was chosen as the minimum energy average lattice parameter, as seen in Table S1 in SM [46], and was performed with an $8 \times 8 \times 8$ supercell (1024 atoms), with periodic boundary conditions. The initial random configuration was annealed at a number of distinct temperatures for 100 sweeps, where one sweep contains 1024 trial swaps. For all simulations below 500 K, the starting configuration was the final configuration from the 500 K Monte Carlo simulation. This was to ensure these low-temperature

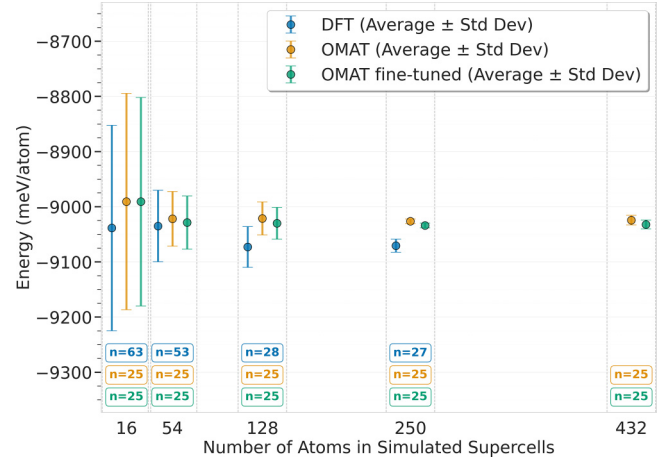


FIG. 5. Convergence of supercell energies for AlTiCrMoW as a function of supercell size, comparing DFT, the OMAT base model, and the fine-tuned OMAT model. Energies are given in meV for supercells from $2 \times 2 \times 2$ to $6 \times 6 \times 6$. DFT values are averaged over all optimized training-set supercells, while OMAT results are from 25 random configurations per size. The number of sampled cells (n) is indicated on the plot.

simulations did not become stuck in a local energy minimum, allowing the systems to be driven towards lower energies. The plots of the Warren-Cowley ASRO parameters and lattice energy versus temperature are shown in Fig. 7. The full sweep-by-sweep evolution of the energy and ASRO parameters for the 500 K simulation is shown in SM [46], where we observe full convergence at 100 sweeps.

Minimal ASRO is present within the cells at high temperatures, although there is a slight preference for Al-Al, W-W, Al-Ti, Cr-Mo, and Mo-W first neighbor pairs as well as W-W second neighbor pairs at temperatures greater than 2000 K. A higher degree of ASRO starts to occur at below 2000 K, where the Ti-Ti, Cr-Cr, Al-W, Ti-W first neighbors and Mo-Mo, Ti-Cr, Al-W, Ti-W, and Ti-Mo second neighbors become disfavored interactions. Likewise, W-W and Al-Ti first neighbors and Ti-Ti second neighbors start to become more favored interactions below 2000 K. At around 1500 K, we see the ASRO curves start to split into their full orderings, where the previously discussed pairs below 2000 K become even more favored or disfavored. At 1500 K, we see different pairs becoming ordered and disordered; Al-Cr first neighbors become disfavored interactions, whereas Cr-W and Ti-Mo first neighbors and W-W, Cr-Cr, Al-Mo second neighbors become favored interactions. These trends continue and become more favored or disfavored as the temperature lowers. From our plots, we predict a change in short-range ordering behavior at $1250 \text{ K} \pm 50 \text{ K}$, and predict an ordered structure at around $250 \text{ K} \pm 25 \text{ K}$. These values were obtained by differentiating the total energy and ASRO curves and locating the peaks. A version of 7 with each unique simulation temperature plotted as a discrete point is available within SM [46]. Figure 8 visualizes the configuration of the AlTiCrMoW supercell at various key temperatures.

In order to compare our results to the base OMAT model, a coarser simulation (fewer discrete temperatures simulated) was performed using the base OMAT model. The resulting

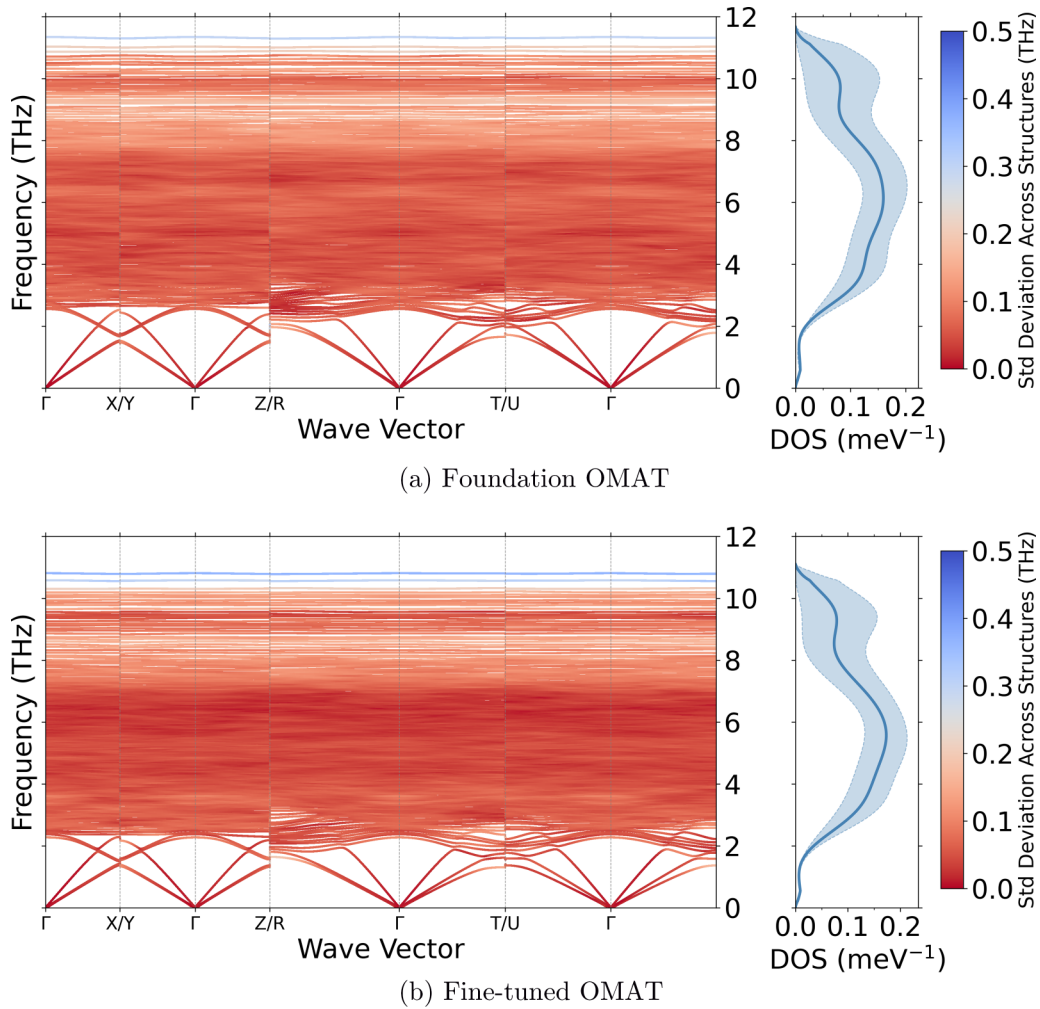


FIG. 6. Phonon dispersion relations (left) and phonon density of states (right) of AlTiCrMoW from the OMAT base model, and the fine-tuned OMAT model. Dispersion curves averaged over five near equiatomic $3 \times 3 \times 3$ (54-atom) supercells constructed so that each element is underrepresented once and the arithmetic average is exactly equiatomic; shaded regions denote configuration-induced standard deviations.

plots of the Warren-Cowley ASRO parameters and lattice energy vs temperature are shown in Fig. 9. A version of Fig. 9 with each unique simulation temperature plotted as a discrete point is available in SM [46].

The simulations with the base OMAT model and the fine-tuned OMAT model predominantly agree. A change in short-range ordering behavior ordering once again occurs around 1250 K, with ordering occurring around 250 K. Most favored/disfavored interactions are similar, with some subtle reranking of preferred interactions. This indicates that fine-tuning the OMAT base model modifies, rather than wholly replaces, the ordering tendencies predicted by the base model, adjusted its predictions for the phase stability of the AlTiCrMoW HEA and enabled simulations like the Monte Carlo workflow presented here to be computationally viable.

To examine whether the fine-tuned and base OMAT models predict different energetically preferred elemental exchanges during Monte Carlo sampling, additional simulations were performed using each model, starting from their respective equilibrated 1500 K configurations. Each simulation was run for 50 Monte Carlo sweeps at 1500 K, followed by a further

50 sweeps at 1 K to sample low-temperature swap energetics. The resulting swap statistics are shown in Fig. 10.

Figure 10 summarizes the energetic response to elemental swaps during Monte Carlo equilibration, providing direct insight into the chemical driving forces underlying ASRO in AlTiCrMoW. Each data point corresponds to the energy change, ΔE , associated with swapping two atoms of different species. Distributions centered near zero indicate that most swap types may be either energetically favorable (negative ΔE , below the dashed line) or unfavorable (positive ΔE , above the line), depending on the local chemical environment.

Clear differences emerge when comparing the two models. The fine-tuned OMAT model exhibits more tightly constrained energy distributions for several swap types and systematically shifted central tendencies for specific elemental pairs. In particular, swaps involving Al display broader and more weakly constrained energy distributions in the base model, whereas the fine-tuned model produces a clearer energetic preference, most notably for the Al–Mo and Al–W pairs. This behavior highlights the bcc-stabilising role of Al once the model is fine-tuned, consistent with established

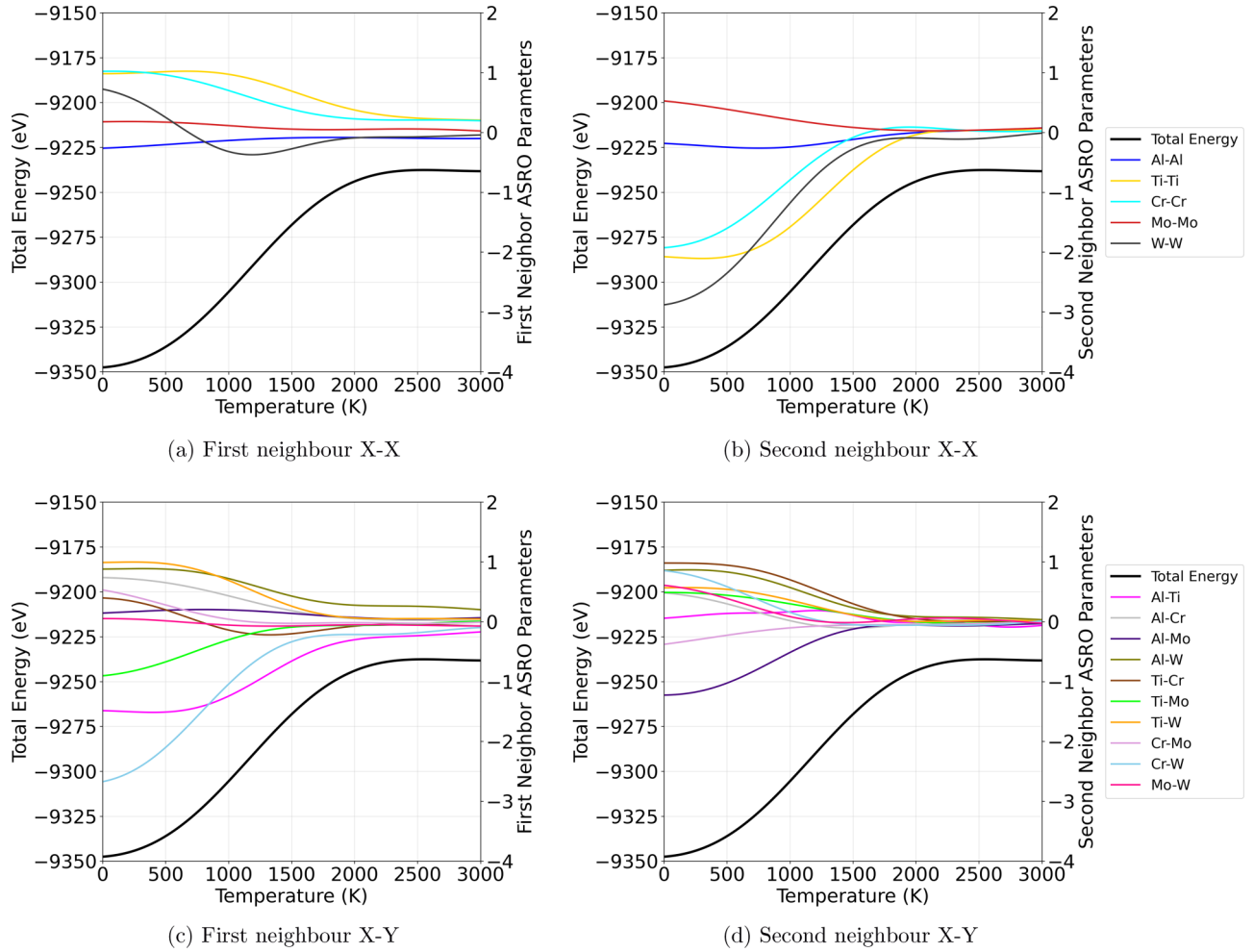


FIG. 7. ASRO and total energy curves for AlTiCrMoW calculated from our MC simulations. X-X represents pairs of the same element, X-Y represents pairs of different elements.

thermodynamic guidelines for refractory and transition-metal-rich bcc alloys (see, e.g., Refs. [88,89]) and with similar observations in CoCrFeMnAl [90], CoCrFeNiAl [18,91], and TiZrHfNbAl systems [88].

In addition, several transition-metal swap types involving Cr, Mo, and W exhibit shifted energetic preferences between the two models. This indirect modification of relative Cr-Mo-W energetics can be viewed as an effective renormalization of the transition-metal interaction landscape, in the sense that fine-tuning on Al-containing environments changes the relative chemical preferences among the transition metals without altering the underlying lattice or symmetry. As a result, the fine-tuned model predicts modified short-range chemical ordering tendencies and corresponding shifts in phase-stability trends, even though both models preserve the overall bcc character of the alloy.

V. VACANCY FORMATION PREDICTIONS IN ALTiCrMoW

Having established the phase stability and short-range ordering behavior of AlTiCrMoW, we now turn to point-defect properties, which govern high-temperature mechanical response through their role in diffusion, creep, and dislocation

climb. Vacancy formation energies and relaxation volumes are central parameters for any theory of atomic transport and time-dependent plasticity [44,92,93]. In compositionally complex alloys, these quantities cannot be assigned a single value: the stochastic disorder ensures that each vacancy sits in a distinct local environment, giving rise to broad distributions of formation enthalpies [94–97] and relaxation volumes [98]. Accessing these distributions requires statistical sampling over many independent atomic configurations [94,96], a demand that places prohibitive strain on first-principles methods when supercells must be large enough to suppress artificial vacancy-vacancy interactions. At the same time, while empirical interatomic potentials offer an inexpensive alternative, their ability to resolve the subtle chemistry of defect energetics in multicomponent alloys remains more limited [99]. The combination of these difficulties has left vacancy properties in HEAs only partially charted, despite their central importance for mechanistic models of high-temperature deformation [100]. The purpose of this analysis is not only to assign a mean vacancy response to each removed species, but to resolve the distribution of relaxation volumes generated by many chemically distinct local environments and to quantify those environmental contributions statistically.

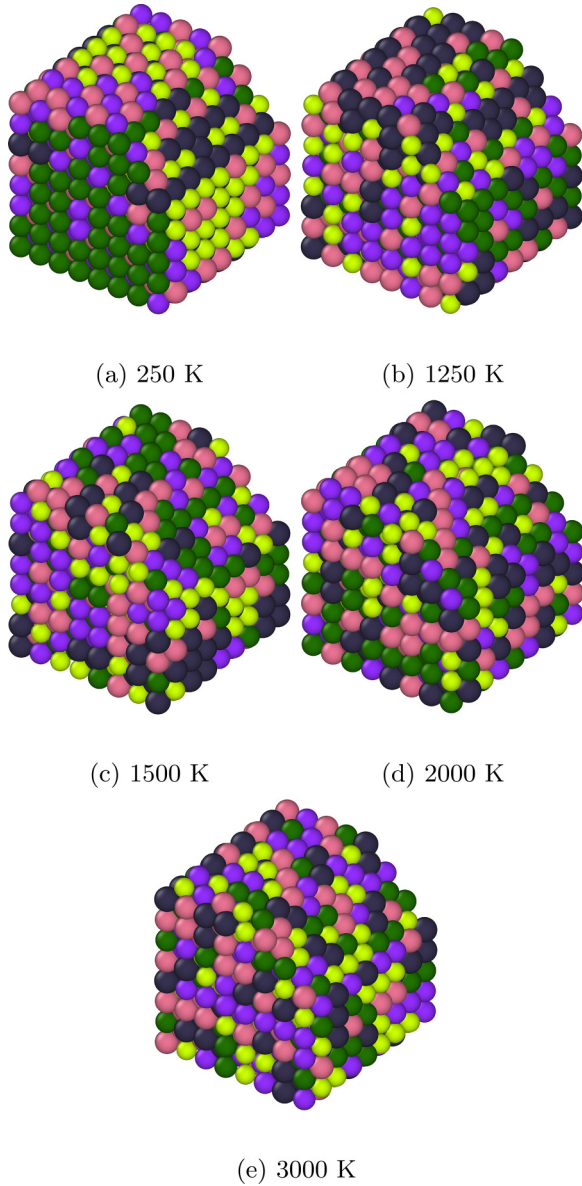


FIG. 8. Visualized configurations from the Monte Carlo simulations for AlTiCrMoW. Al, Ti, Cr, Mo, and W are colored blue, green, yellow, pink, and navy blue, respectively. A slice has been made through the simulation cell to more clearly visualize the ASRO present. Images generated using OVITO [87].

The MLIP framework developed here offers a solution. With near-DFT accuracy but orders-of-magnitude lower computational cost, we can simulate ensembles of large disordered supercells and extract statistically meaningful distributions of defect properties. Moreover, the chemical specificity encoded in the MLIP enables us to correlate vacancy behavior with local composition, revealing which elemental neighborhoods promote or suppress vacancy formation and relaxation. This capability is particularly valuable for refractory HEAs intended for service above half the melting temperature, where vacancy-mediated processes dominate mechanical degradation [45].

To probe the defect thermodynamics of complex concentrated alloys, we have employed our fine-tuned MACE

potential to investigate vacancy formation and relaxation volumes in AlTiCrMoW. We generated a series of randomly selected $5 \times 5 \times 5$ equimolar supercells, corresponding to 250 atoms, and introduced single vacancies of different elemental types. The motivation for this study is twofold. First, the relaxation volume Ω is the primary elastic descriptor of a point defect: it quantifies the volumetric response of the lattice to defect formation, directly controls the long-range elastic interactions with dislocations, and enters continuum models of diffusion [44] and creep [45]. Second, in HEAs, where the local environment varies stochastically, Ω is not a fixed number but a distribution reflecting the chemical complexity of the alloy. Understanding how local composition biases vacancy contraction or expansion is thus essential for linking atomistic defect energetics to macroscopic transport and mechanical properties.

A direct treatment of these effects at the DFT level can be prohibitively difficult. Vacancies in HEAs are highly non-symmetric, requiring supercells of several hundred atoms in order to eliminate spurious image interactions [92,100]. Even with such large cells, the vacancy field couples to its periodic replicas, producing residual vacancy-like displacements at the boundaries that are smeared by periodicity, leading to Pulay-like oscillations as the ionic relaxations interact with the electronic sector [92]. The stochastic compositional stresses inherent to a disordered lattice [94,95] complicate matters further, since the stochastic local chemistry of a disordered lattice also complicates the extraction of defect-induced force fields, since the reference and defective configurations must be compared at a common geometric state in order to separate the vacancy perturbation from environment-specific bonding and internal stress contributions. [101]. Moreover, the breaking of local symmetry around the vacancy amplifies k -point sampling sensitivity [96], and the broad distribution of relaxation patterns requires averaging over many independent realizations [94,96,97]. These challenges combine to make systematic DFT sampling of vacancy relaxation volumes in HEAs highly computationally intensive, and limit its current predictive ability. By contrast, the MACE potential allows us to simulate large disordered supercells at low cost while retaining near-DFT accuracy in the force response. This enables us to assemble statistically meaningful datasets across many random environments and to resolve how local composition governs the vacancy relaxation volume.

A. Vacancy formation energies

The vacancy formation energy is defined as the energy required to remove an atom from the perfect crystal. In practice, for a supercell containing N atoms, this is computed as:

$$E_f = E_{\text{defect}}(N-1) - \frac{N-1}{N} \times E_{\text{perfect}}(N), \quad (22)$$

where $E_{\text{defect}}(N-1)$ is the total energy of the relaxed supercell with one vacancy (containing $N-1$ atoms), and $E_{\text{perfect}}(N)$ is the total energy of the perfect supercell containing N atoms. The factor $(N-1)/N$ accounts for the chemical

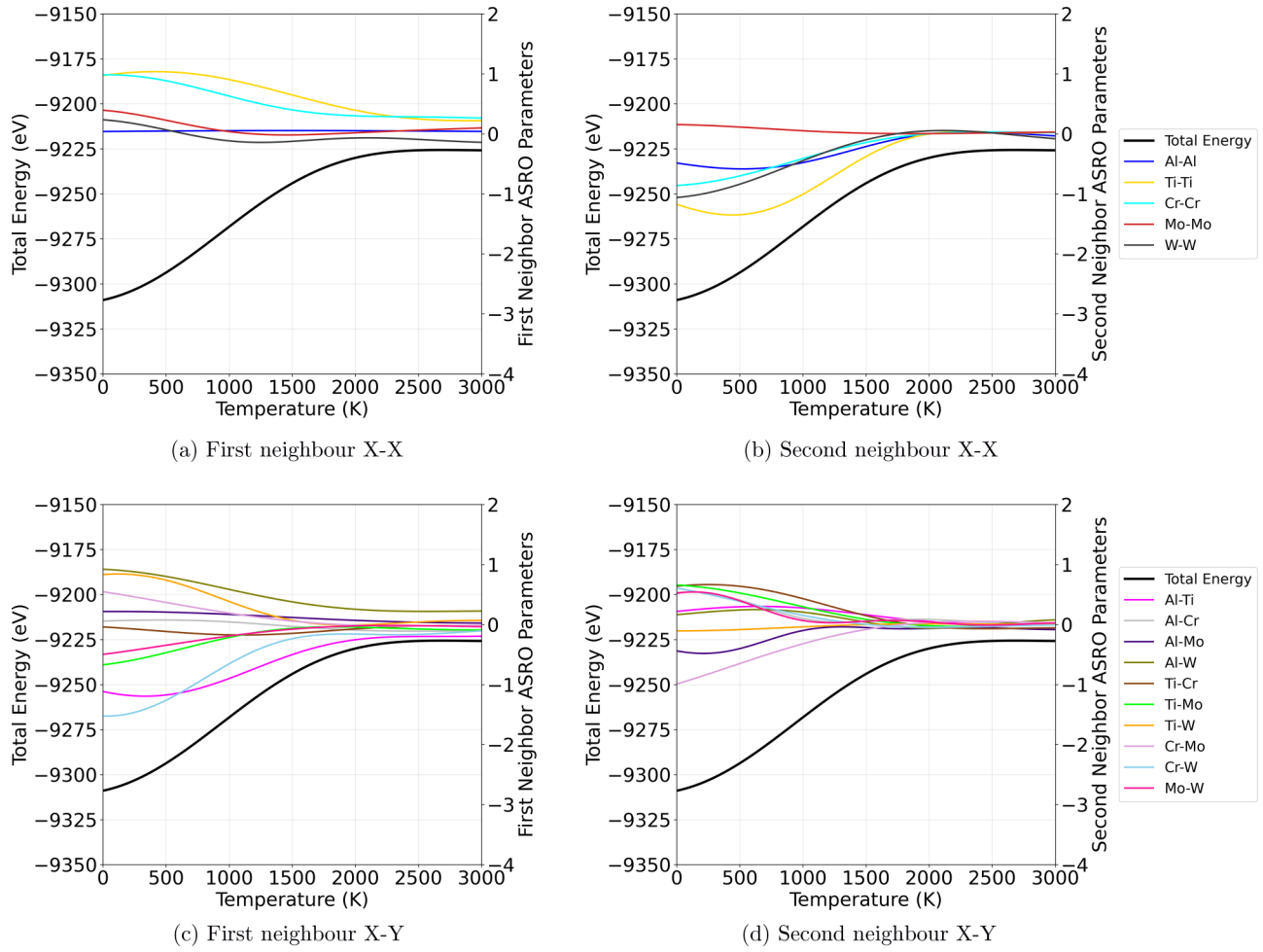


FIG. 9. ASRO and total energy curves for AlTiCrMoW calculated from our MC simulations using the base OMAT model. X-X represents pairs of the same element, X-Y represents pairs of different elements.

potential of the removed atom, ensuring that the formation energy represents the cost of creating the vacancy. All energies were obtained following the annealing-quench-minimization protocol described in Sec. III A.

B. Kanzaki force formalism for relaxation volumes in HEAs

In the case of HEAs, the relaxation volume associated with a point defect can be obtained most directly from the defect-induced Kanzaki forces and the dipole moment tensor.

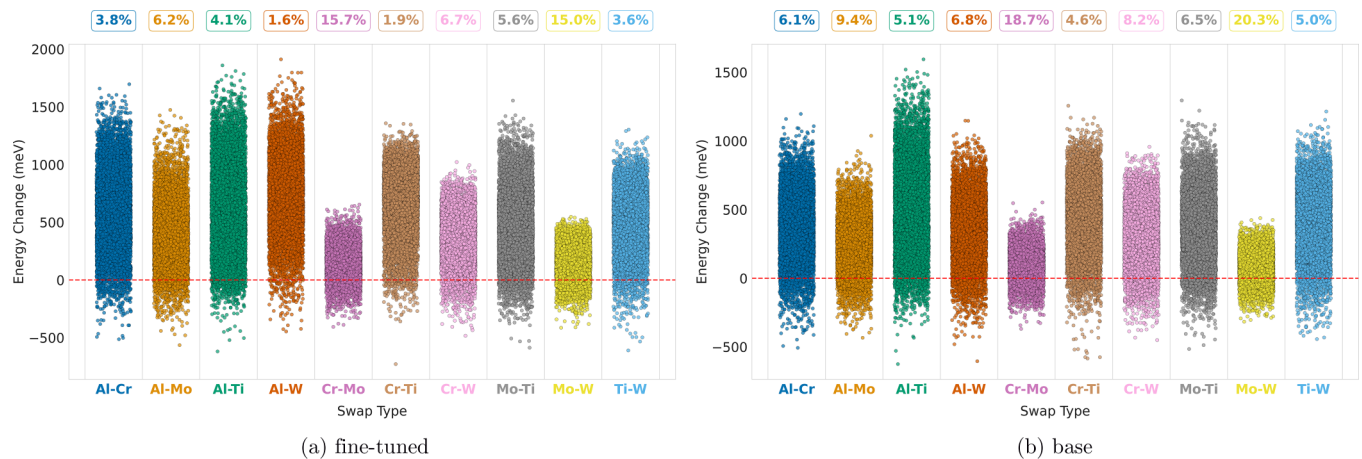


FIG. 10. The swap statistics for the (a) fine-tuned OMAT model MC simulations and (b) base OMAT model MC simulations.

In the Kanzaki force formalism [102], one defines the force distribution $\{\mathbf{F}_i^K\}$ as the set of forces which, when applied to the perfect reference lattice, reproduce the displacements observed in the defective configuration. Operationally, we determine these forces from the difference

$$\mathbf{F}_i^K = \mathbf{F}_i^{\text{def}} - \mathbf{F}_i^{\text{perf}}, \quad (23)$$

where $\mathbf{F}_i^{\text{def}}$ are the Hellmann–Feynman forces on atoms i in the supercell containing the defect, and $\mathbf{F}_i^{\text{perf}}$ are those for the same supercell in the perfect crystal. The subtraction is crucial in a HEA, because chemical disorder produces strong spatial heterogeneity in local bonding and internal stress fields (see, e.g., Refs. [4,101,103]). To isolate the Kanzaki force distribution associated with introducing a vacancy, we therefore subtract the reference force field of the corresponding chemically identical, vacancy-free configuration evaluated at the same geometric reference as the defect calculation. This removes large, environment-specific background contributions and leaves only the incremental force response induced by the vacancy.

In practice, we computed the Kanzaki force distributions using the annealing-minimization protocol described in Sec. II D for the training set. We thus analysed 34 $5 \times 5 \times 5$ fully relaxed perfect supercells. The reference forces $\{\mathbf{F}_i^{\text{perf}}\}$ are evaluated for the chemically identical vacancy free configuration at the same geometric reference used for the defect calculation, so that Eq. (23) isolates the incremental force field generated by removing the atom.

To generate the defect, a single atom was removed at random from the supercell to form a vacancy. The defective configuration was again equilibrated with the identical annealing and minimization protocol, ensuring a consistent thermal history between perfect and defective structures. Finally, to extract the Kanzaki forces, the removed atom was reinserted at its original lattice site into the relaxed defective cell, the atomic positions were frozen, and a single-point force calculation was performed [65,104]. The resulting forces $\{\mathbf{F}_i^{\text{def}}\}$ correspond to those exerted by the relaxed defective lattice on the reinstated atom, and their difference from the perfect forces yields the Kanzaki forces via Eq. (23). This procedure ensures that the extracted $\mathbf{F}_i^K$ represent only the defect-induced perturbation, disentangled from the stochastic compositional stress fields inherent to the disordered HEA matrix.

The Kanzaki forces were then summarized by their elastic dipole moment tensor [65,105,106],

$$P_{ij} = \sum_a R_i^{(a)} F_j^K(a), \quad (24)$$

where $\mathbf{R}^{(a)}$ is the position vector of atom a in the perfect lattice relative to the defect center. The tensor P_{ij} couples linearly to the applied strain in the standard elastic-dipole formalism [107], and its trace gives access to the relaxation volume [106,108]

$$\Omega = \frac{1}{3K} \text{Tr } P, \quad (25)$$

where K is the bulk modulus of the alloy, taken from the calculation of the same random supercell as described in Sec. III. In practice, we average Ω across multiple independent HEA

realizations in order to sample the distribution of local environments.

It is important to note that this Kanzaki-based route is considerably simpler in disordered alloys than the alternative strain–vertex approach (see, e.g., Refs. [109,110]). In the latter method, one measures the change of defect formation energy $E_f(\boldsymbol{\epsilon})$ under imposed hydrostatic strain ϵ_h and extracts the relaxation volume from

$$\Omega = - \left. \frac{\partial E_f}{\partial (3K\epsilon_h)} \right|_{\epsilon_h=0}. \quad (26)$$

However, in HEAs, the reference $\epsilon_h = 0$ state is ill-defined, since the disordered lattice carries an intrinsic residual stress. One must first determine the strain $\boldsymbol{\epsilon}_0$ which minimizes the total energy of the defect-free supercell and then rereference all derivatives to this $\boldsymbol{\epsilon}_0$. This additional step significantly complicates the analysis and introduces additional numerical uncertainty, particularly because the residual stress varies from one random configuration to the next. For this reason, here we adopt the Kanzaki force formalism, which isolates the defect contribution directly via the force difference in Eq. (23) and requires no prior determination of a stress-free strain state, and use the strain vertex approach to validate some of the unusual results we report in the sequel. These vacancy statistics are directly relevant to diffusion, creep and irradiation response, as their relaxation volume encodes the elastic dipole of the vacancy, which modulates migration barriers under local stress and thus affects vacancy hopping rates and the resulting diffusion behavior [44].

C. Dipole moment tensors, relaxation volumes, and statistical analysis of the role of the local environment

The results for the dipole moment tensor calculations show that vacancy formation in AlTiCrMoW produces strongly element-dependent dipole tensors and relaxation volumes, reflecting distinct local elastic responses for each species. These values are summarized in Table IV, with the corresponding relaxation volumes illustrated in Fig. 11.

Chromium vacancies exhibit the largest positive relaxation volume ($\Omega_{\text{Cr}} = 3.82 \pm 0.93 \text{ \AA}^3$), indicating substantial outward relaxation of the surrounding atomic structure. This behavior suggests that Cr atoms experience significant compressive stress in the bcc HEA matrix, consistent with its smaller atomic radius compared to the average alloy composition. Once the chromium atom is removed, the volume of the vacancy expands to relieve the strain on the overall bcc structure.

In contrast, aluminium, titanium, molybdenum and tungsten vacancies all show average negative relaxation volumes ($\Omega_{\text{Al}} = -0.85 \pm 0.70 \text{ \AA}^3$, $\Omega_{\text{Ti}} = -1.98 \pm 1.12 \text{ \AA}^3$, $\Omega_{\text{Mo}} = -0.43 \pm 1.13 \text{ \AA}^3$, and $\Omega_{\text{W}} = -1.14 \pm 1.29 \text{ \AA}^3$), indicating inward relaxation upon vacancy formation. This is the expected behavior of the relaxation of a point defect in a metallic system. These calculated behaviors of point defects are physically plausible, as a chromium atom has an ionic radius of 128 pm, whilst the other four metals all have ionic radii between 130 and 140 pm. To ensure the overall bcc structure is stable, each lattice point will need to occupy a volume which is

TABLE IV. Element-averaged dipole tensors for different vacancy types (averaged over 34 structures each).

Vacancy type	Mean dipole tensor (eV)		
Al	-0.925 ± 1.402	-0.084 ± 1.325	-0.107 ± 1.627
	-0.084 ± 1.325	-1.111 ± 1.188	0.421 ± 1.694
	-0.107 ± 1.627	0.421 ± 1.694	-0.904 ± 1.303
Ti	-2.468 ± 1.662	0.266 ± 1.465	0.036 ± 1.348
	0.266 ± 1.465	-2.246 ± 1.804	-0.409 ± 1.311
	0.036 ± 1.348	-0.409 ± 1.311	-2.160 ± 1.443
Cr	4.191 ± 1.534	0.038 ± 0.786	-0.082 ± 0.921
	0.038 ± 0.786	4.748 ± 1.391	-0.070 ± 0.831
	-0.082 ± 0.921	-0.070 ± 0.831	4.333 ± 1.488
Mo	-0.500 ± 1.549	-0.516 ± 1.109	0.182 ± 1.072
	-0.516 ± 1.109	-0.602 ± 1.445	-0.024 ± 1.109
	0.182 ± 1.072	-0.024 ± 1.109	-0.383 ± 2.109
W	-1.573 ± 1.766	0.353 ± 1.605	0.026 ± 1.549
	0.353 ± 1.605	-1.056 ± 1.815	-0.145 ± 1.433
	0.026 ± 1.549	-0.145 ± 1.433	-1.331 ± 2.041

around the average ionic radius, hence the observed expansion with a chromium vacancy.

The magnitude of relaxation volumes (ranging from -1.1 to $+3.6 \text{ \AA}^3$) indicates significant local distortions around vacancies, which may influence diffusion pathways, precipitation behavior, and mechanical properties in this HEA.

The dispersion of relaxation volumes around these averages, appreciable in Fig. 11, is large, reflecting the effect the heterogeneous local environments has in vacancies in HEAs. To study and quantify the effect of local composition, we parametrized the environment around each vacancy by computing a distance-weighted elemental composition

$$\phi_s^{(i)} = \frac{\sum_{k \in \mathcal{V}_s(i)} w_k}{\sum_{k \in \mathcal{S}(i)} w_k}, \quad w_k = r_k^{-2}, \quad (27)$$

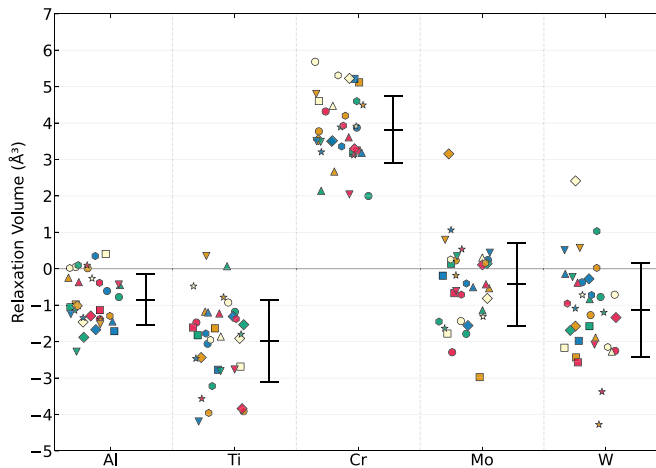


FIG. 11. Vacancy relaxation volumes in AlTiCrMoW from MACE calculations on $5 \times 5 \times 5$ random supercells. Each point corresponds to a single vacancy site.

where i is the vacancy site, $\phi_s^{(i)}$ denotes the effective fraction of species $s \in \{\text{Al, Ti, Cr, Mo, W}\}$ in the first two coordination shells about i , $\mathcal{S}(i)$ is the full neighbor set of site i , $\mathcal{V}_s(i)$ is the set of neighboring atoms of type s about the site i , and w_k are inverse-square weights reflecting the point defect's displacement field scaling with r^{-2} [106]. Periodic boundary conditions were enforced to avoid truncation artefacts. The resulting distance-weighted composition vectors $\phi_i = (\phi_{\text{Al}}^{(i)}, \phi_{\text{Ti}}^{(i)}, \phi_{\text{Cr}}^{(i)}, \phi_{\text{Mo}}^{(i)}, \phi_{\text{W}}^{(i)})$ provide a compact, continuous representation of the local environment. By construction, $\sum_s \phi_s^{(i)} = 1$ for each vacancy site i .

We then modelled the dependence of the relaxation volume on ϕ_i using mixed regression models with a sum-to-zero constraint on the elemental coefficients, such that $\sum_s \beta_s = 0$. This ensures that β_s is interpreted as the deviation relative to the mean environment. Three responses were considered: (i) the sign of the relaxation volume, for which we used a logistic regression

$$\text{logit Pr}[\Omega_i > 0 \mid \phi_i] = \alpha + \sum_s \beta_s \phi_s^{(i)} + u_{j(i)}, \quad (28)$$

where α is the baseline log-odds, ϕ_i is the local composition vector for vacancy site i , $\phi_s^{(i)}$ the distance-weighted fraction of species s in its first two coordination shells [see Eq. (27)], which is treated purely as a compositional predictor rather than a physical alteration of site occupancy, and $u_{j(i)} \sim \mathcal{N}(0, \tau^2)$ a random intercept accounting for structure-to-structure offsets, where $j(i)$ denotes the index of the parent supercell (configuration) containing vacancy site i and $\mathcal{N}(0, \tau^2)$ is a normal distribution with mean zero and variance τ^2 ; (ii) the magnitude of Ω_i , fitted by a linear model with the same predictors; and (iii) the formation energy, likewise treated linearly. The coefficients β_s thus quantify the effect of relative enrichment in element s within the local shell, i.e., changes in $\phi_s^{(i)}$ at fixed $\sum_s \phi_s^{(i)} = 1$, and are interpreted as deviations from the mean environment under the sum-to-zero constraint.

These statistical models allow us to isolate the influence of each species in the local coordination shell. Figure 12 shows the fixed effect coefficients from the sum-to-zero constrained regression. It shows the effect of each element in the local environment: Al and Cr neighbors bias strongly towards contraction (negative β), Mo and W towards expansion (positive β), and Ti lies between these extremes. This makes physical sense, as Cr in particular has the smallest atomic radius in the quinary alloy, and W the largest, so they are bound, respectively, to promote a contraction and expansion of their own local environments.

Indeed, these plots confirm that the presence of increasing amounts of Al and chromium atoms within the first two coordination shells promotes volume reduction of a vacancy, whereas the presence of increasing amounts of Ti, Mo, and W within the first two coordination shells promotes volume expansion. We also note that an increasing presence of W atoms within the first two coordination shells has a major effect on the vacancy formation energy, where an increase in tungsten concentration provides a stabilising effect. Therefore we predict vacancies are most likely to form within regions of a given alloy composition where tungsten - tungsten SRO

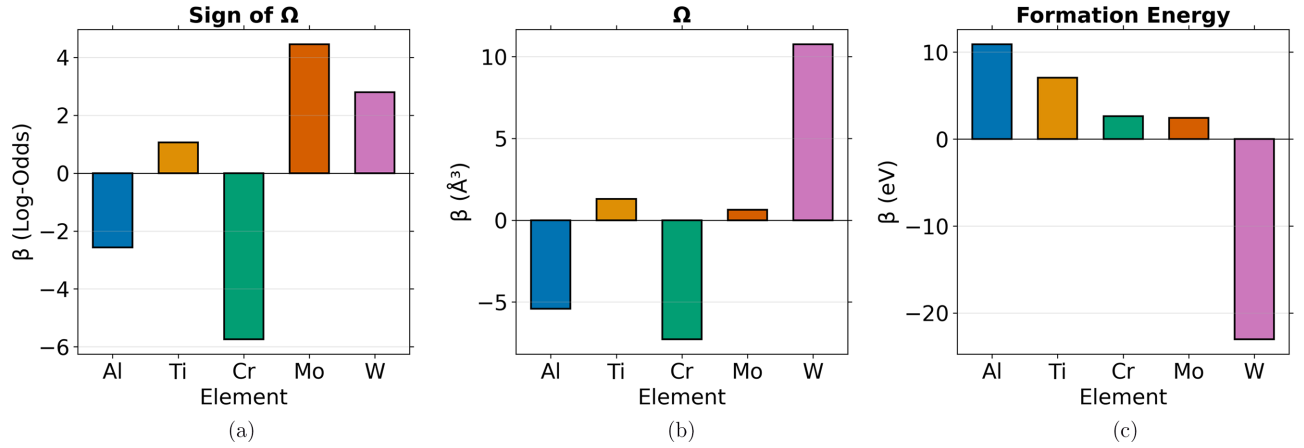


FIG. 12. Statistical analysis for vacancy formation in AlTiCrMoW.

occurs. Further statistical analysis is provided within Sec. VIII of SM [46]. For one of the structures, we compared the MACE vacancy formation energies to the respective DFT formation energies and found that DFT on average calculates the vacancy formation energy to be roughly 2.5 eV higher on average for each elemental vacancy. This was an expected result due to the previously discussed difficulties with modeling HEA vacancies via DFT.

To clarify the hierarchy of elemental effects, Fig. 13 shows the pairwise contrasts $\beta_i - \beta_j$. These values express how much more strongly species i biases the vacancy response compared to species j , independent of any arbitrary scale. For the sign of the relaxation volume (left panel), the heatmap shows that a Mo-rich local shell increases the odds of expansion by more than one decade relative to Cr, while W similarly favors expansion compared to both Al and Ti. By contrast, Cr consistently appears as the most contraction-prone neighbor, with $\beta_{\text{Cr}} - \beta_{\text{Mo}} = -10.3$ and $\beta_{\text{Cr}} - \beta_{\text{W}} = -8.8$. The magnitude and energy panels (middle and right) reinforce the same ordering: W and Mo promote larger relaxation volumes, Cr drives strong contraction, and Al and Ti remain intermediate. These pairwise comparisons are especially valuable because they distill the regression into physically transparent relative rankings, allowing us to state not just whether a species tends

to expand or contract a vacancy, but by how much more or less it does so compared to another species.

Together, these plots demonstrate that the large dispersion of vacancy relaxation volumes in the raw data is not random, but reflects systematic, element-specific contributions from the immediate coordination environment. The coefficient bar chart (Fig. 12) provides a species-resolved measure of the effect, while the pairwise contrasts (Fig. 13) establish the relative ordering across all species. This dual presentation provides quantitative insight into the local stress states experienced by different atomic species in the bcc AlTiCrMoW matrix. The large positive value for Cr and negative values for Al and Ti are consistent with atomic size mismatch effects, where smaller atoms (Cr) are compressed while larger atoms (Al, Ti) are under tension in the average bcc environment.

These results show that vacancy relaxation in HEAs is not a uniform property but a strongly environment-dependent phenomenon. The sign and magnitude of the relaxation volume, and by extension the local strain field of the defect, depend systematically on the chemical makeup of the nearest neighbor shell, and these can be quantified and systematically studied statistically via the MLIP-based methodology here presented. Crucially, transport coefficients derived from vacancy mechanisms, such as diffusivity [93] or creep rates [44],

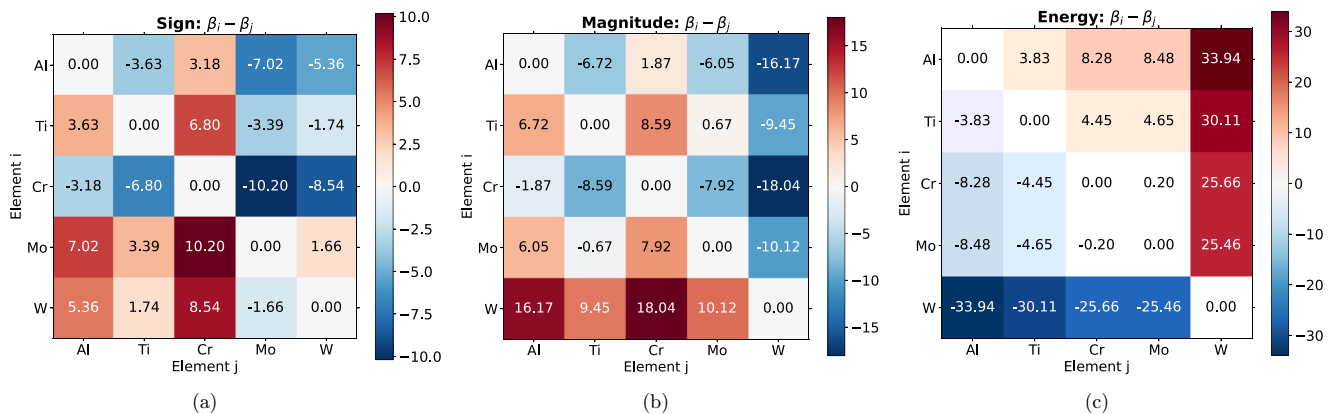


FIG. 13. Pairwise differences of logistic regression coefficients, $\beta_i - \beta_j$, for the probability that a vacancy exhibits positive relaxation volume. Positive values indicate that enrichment in species i relative to species j increases the odds of $\Omega > 0$, while negative values indicate the opposite. Coefficients are sum-to-zero coded, so differences quantify relative rather than absolute species effects.

will inherit a broad distribution rather than a single effective value. It also suggests that alloy design strategies which tune the relative abundance of Cr, W, or Mo in local environments can be expected to alter vacancy stability and mobility in predictable ways. More generally, the ability of a statistical framework to disentangle these element-specific contributions shows that vacancy behavior in compositionally complex alloys can be treated not as random noise but as a structured response of the lattice to local chemistry.

VI. CONCLUSIONS

In summary, we have developed and validated an MLIP for the prototypical refractory HEA AlTiCrMoW that achieves near DFT accuracy while remaining orders of magnitude more efficient. The fine-tuned MACE model reproduces lattice parameters, volumes, elastic properties, and phonon spectra with high fidelity, demonstrating robust transferability across the relevant configurational space. This establishes a reliable atomistic tool for probing the thermodynamics of chemically complex alloys beyond the reach of direct DFT. If another alloy contains some similarities in composition space, the model we trained would be able to be fine-tuned further with a smaller, more focused training set of the alloy.

By using the MACE potential within Metropolis Monte Carlo simulations, we accessed equilibrium ASRO and predicted the ordering transition temperature that defines the ordering transition of AlTiCrMoW. The results reveal both the temperature range of stability and the specific elemental associations that persist across temperatures, thereby providing an atomistic basis for understanding the strengthening behavior of concentrated refractory alloys.

The framework presented here, combining KKR-CPA effective medium theory, a perturbative concentration wave analysis, and targeted fine-tuning of MACE, constitutes a general strategy for constructing system-specific MLIPs when experimental data are scarce, and brute-force DFT sampling is infeasible. It is directly transferable to other refractory HEAs and emerging multicomponent systems where complex chemical interactions and extreme service conditions challenge conventional modeling.

A key demonstration of this capability is the quantitative resolution of vacancy formation and relaxation behavior in such a chemically complex refractory alloy as AlTiCrMoW, enabled by the accuracy and scalability of the MACE potential. Although the qualitative role of the local environment on defect energetics in HEAs has been recognized [94,99], a systematic and statistically converged characterization of how specific coordination-shell compositions modulate relaxation volumes has not been reported. Conventional EAMs lack the chemical resolution to capture charge transfer and anisotropic distortions, while DFT is computationally prohibitive for the hundreds of large disordered supercells needed to sample the full configurational ensemble. The present MACE model bridges this gap by providing near-DFT fidelity in forces and energies at minimal cost, allowing direct computation of Kanzaki forces, dipole tensors, and relaxation volumes across many random environments. The resulting statistics reveal that the volumetric response of a vacancy is highly sensitive to its immediate neighborhood: enrichment in W

and Ti promotes expansion, while Cr and Al favor contraction, establishing a clear hierarchy of local elastic responses ($W > Ti > Mo > Al > Cr$).

These findings show that vacancy relaxation in HEAs is not a single-valued material constant but a distribution governed by the chemical topology of the local shell. Such quantitative mapping of defect-elastic coupling opens a path towards predictive models of mass transport, segregation, and creep in compositionally complex alloys, where the statistical mechanics of local chemical environments must be treated explicitly. The same framework can be extended to study solute diffusion, dislocation climb, and irradiation-driven defect production, providing a unified route to connect atomistic thermodynamics with macroscopic high-temperature performance.

Looking forward, the efficiency of the present MLIP enables simulations at scales sufficient to capture extended defects, grain boundaries, and deformation mechanisms, bringing into reach atomistic studies of strengthening, creep, and phase stability in industrially relevant environments. Incorporating finite-temperature DFT training data and extending to off-stoichiometric and oxidising conditions will further broaden predictive capability. In this way, machine-learned potentials offer a practical route to accelerating the discovery and optimization of next-generation structural alloys for high-temperature, corrosive, and mechanically demanding applications.

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DATA AVAILABILITY

The fine-tuned OMAT model is available on GitHub [111]. Any other data can be provided upon request.

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