

Supplemental Materials to the article: Impact of local lattice relaxations on phase stability and chemical ordering in bcc NbMoTaW high-entropy alloys explored by ab initio based machine-learning potentials

Tatiana Kostiuchenko, Fritz Körmann, Jörg Neugebauer, Alexander Shapeev

1 LRP predictive power analysis

The LRP obtained by our adaptive training procedure predicted three phases: the disordered, semi-ordered, and ordered one. Disordered and semi-ordered structures are not a part of the training set. Therefore we analyze how well LRP reproduces these phases.

To that end, we took the ordered structure of eight $\langle 100 \rangle$ -layers of atoms in the following order: Nb-Mo-Ta-W-W-Ta-Mo-Nb, the semi-ordered structures consist of 13 $\langle 100 \rangle$ -layers, Nb-Mo-Ta-W-W-Ta-Mo-Ta-W-W-Ta-Mo-Nb and W-Ta-Mo-Nb-Nb-Mo-Ta-Mo-Nb-Nb-Mo-Ta-W, and 10 $4 \times 4 \times 4$ disordered supercell structures generated by MC at $T=400\text{K}$. We calculated the difference between the energies predicted by LRP and obtained via DFT that both take local lattice distortions into account. The results are presented in Table 1. It is not surprising that the ordered ground state is reproduced with very high accuracy since it and similar structures are added to the training set by our training procedure. Large disordered structures generated by MC at $T = 400\text{K}$ are reproduced with the accuracy of 0.4 meV/atom —this indicates that the $4 \times 2 \times 2$ training samples well-represent the disordered phase. The semi-ordered phase is the most challenging for the description by LRP, the respective structures consisted of 13 $\langle 100 \rangle$ -layers are reproduced with the

Phase	Error (meV/atom)
ordered	0.1
semi-ordered	1.7
disordered 4x4x4	0.4

Table 1: The error of the LRP prediction for the various discovered phases. The error of the disordered and semi-ordered phases were computed by averaging over the representative configurations. The semi-ordered structures are not commensurate with the cell size of the training configurations (4x2x2 and 2x2x2)—hence the largest error. Nevertheless, the predictive error is still within the 95% confidence interval. We attribute it to the fact that although LRPs were not trained on similar configurations, they still have “seen” similar atomic environments in the training configurations.

error of 1.7 meV/atom. It is important to note, that the semi-disordered phase is not equiatomic, while our training set contains mainly equiatomic concentrations of components. In addition, the semi-ordered structures are not commensurate with the cell size of the configurations in the training set. Nevertheless the 1.7 meV/atom error is still within the confidence interval of our model assuming a normal distribution of error with a root-mean-square of 1 meV/atom. We attribute the low prediction error of LRP to the fact that the LRP still “saw” the relevant atomic environments in the training set.

2 Analysis of importance of relaxation effects

For a better understanding of the effect of local lattice distortions on the stability of phases, we analyze the extent in which the relaxation changes the energy. To that end, we took representative structures of the phases arising in both unrelaxed and relaxed scenarios and relax them with DFT. Namely, we took 10 B2(MoW;NbTa) 2x2x2 structures, B2(Mo;Ta) and B32(Nb;W) structures, one ordered ground-state structure consisted of eight $\langle 100 \rangle$ -layers of atoms in the following order: Nb-Mo-Ta-W-W-Ta-Mo-Nb, the semi-ordered structures consisted of 13 $\langle 100 \rangle$ -layers of atoms, Nb-Mo-Ta-W-W-Ta-Mo-Ta-W-W-Ta-Mo-Nb and W-Ta-Mo-Nb-Nb-Mo-Ta-Mo-Nb-Nb-Mo-Ta-W, and finally 10 disordered structures generated by running MC at T=400K. We compared the energies of these structures before the local lattice relaxation and after. The results of comparison are shown in Table 2. For the structures obtained in the unrelaxed scenario the relaxation effects are insignificant. Indeed, B2(Mo;Ta) and B32(Nb;W) are already at the energy minimum thanks to their symmetry. The average energy decrease for B2(MoW;NbTa) structures is only 0.1 meV/atom. In contrast, the energies of the structures described in the relaxed scenario decrease

42 significantly. The energy of the ordered structure decreased by 16.7 meV/atom after relaxation.
43 The average energy decrease in the case of semi-ordered structures is 15.8 meV/atom. The aver-
44 age energy decrease for large disordered structures is 13.3 meV. This illustrates the importance of
45 taking into account local lattice relaxation in predicting the stability of HEAs.

Phase	ΔE (meV/atom)
disordered 4x4x4	-13.3
semi-ordered	-15.8
ordered	-16.7
B2(Mo,W;Ta,Nb)	-0.1
B2(Mo;Ta)	0.0
B32(Nb;W)	0.0

Table 2: The DFT-computed energy differences before and after local relaxation for the discovered various phases. The energy differences were obtained by averaging over the structures representing the phases. The results explain the importance of local lattice distortions: only by introducing them we managed to find the phases that otherwise a Monte-Carlo algorithm would not be able to predict.