

SUPPLEMENTARY INFORMATION

Specialising neural network potentials for accurate properties and application to the mechanical response of titanium

Tongqi Wen^{1,2}, Rui Wang¹, Lingyu Zhu¹, Linfeng Zhang³,
Han Wang^{4,*}, David J. Srolovitz^{5,6}, and Zhaoxuan Wu^{1†}

¹Department of Materials Science and Engineering, City University of Hong Kong, Kowloon, Hong Kong SAR, China

²Hong Kong Institute for Advanced Study, City University of Hong Kong, Kowloon, Hong Kong SAR, China

³Beijing Institute of Big Data Research, Beijing, China

⁴Laboratory of Computational Physics, Institute of Applied Physics and Computational Mathematics, Beijing, China

⁵Department of Mechanical Engineering, The University of Hong Kong, Hong Kong SAR, China

⁶International Digital Economy Academy (IDEA), Shenzhen, China

I. SUPPLEMENTARY METHODS

Initialisation Step

In the Initialisation step, the uniform volume scaling factor is from 0.94 to 1.06 with a step size of 0.02. Random perturbations are applied to the atomic coordinates by randomly adding displacements uniformly distributed within $\pm 0.01\text{\AA}$. In addition, perturbations are added to the super cell vectors using a symmetric deformation matrix with components chosen at random from a uniform distribution of ± 0.03 . The *ab initio* molecular dynamics (AIMD) simulations were performed at 100 K with all DFT settings the same as that of the other DFT calculations (see the Methods section in the manuscript).

DP-GEN Loop Step

Training an ensemble of trial Deep Potentials (DPs). The sizes of the embedding and fitting nets are the same as those in the training process in Specialisation step (see the Methods section in the manuscript). The cutoff of the DP smooth edition model is $6.0\text{\AA}$ here. The learning rate starts from 1×10^{-3} and ends at 5×10^{-8} after 4×10^5 training steps. An ensemble of four models are trained on the same training sets with the same neural network architecture but starting from different random seeds. In addition, the prefactors of the energy, atomic force and virial tensor in the loss functions are $p_e^{\text{start}}=0.02$, $p_e^{\text{limit}}=2$, $p_f^{\text{start}}=1000$, $p_f^{\text{limit}}=1$, $p_v^{\text{start}}=0$, and $p_v^{\text{limit}}=0$, respectively.

Classical MD based on an ensemble of trial DPs. The DP-based MD (DPMD) is performed using the LAMMPS package¹ to explore either bulk and surface structures. Temperatures are controlled via a Nosé-Hoover thermostat and barostat. Initial super cells of $2 \times 2 \times 2$ are created based on randomly perturbed HCP, BCC and FCC primitive cells. For simulations in the 50 to 1746.9 K (90% of the Ti melting point) temperature range, MD simulations are performed

for an isothermal-isobaric (*NPT*) ensemble where the simulation box shape and size vary independently and pressures are varied from 0.001 to 50 kBar. For simulations in the 1941 to 3687.9 K (190% of the Ti melting point) temperature range, MD simulations are performed using the *NPT* ensemble with fixed box-shape over the same range of pressures. In each iteration, simulations are performed at 5 temperatures and 8 pressures within the specified ranges for a total of 40 MD simulation conditions. Each iteration starts from the lowest temperature and then is slowly ramped to the simulation temperature of interest at fixed pressure (i.e., 0.001, 0.01, 0.1, 1, 5, 10, 20, 50 kBar). The initial surface structures are constructed by extending the supercell of the HCP, BCC and FCC structures along specified crystallographic directions ($\{100\}$, $\{110\}$, $\{111\}$ in BCC, $\{100\}$, $\{110\}$, $\{111\}$ in FCC and $\{0001\}$, $\{10\bar{1}0\}$, $\{10\bar{1}1\}$, $\{11\bar{2}2\}$ in HCP, respectively) and terminated with a vacuum layer of $> 10\text{\AA}$. Surface super cells are then scaled from 0.96 to 1.06 with a step of 0.02. The surface configurations in DPMD were simulated within a canonical (*NVT*) ensemble in the 50 to 3687.9 K temperature range. The lower and upper bounds of $\sigma\{\mathbf{f}_i^\alpha\}$ (see Fig. 1 in the manuscript) are 0.10 and 0.25 from 0 to 2717.4 K and from 0.15 and 0.30 for 2911.5 to 3687.9 K for exploring bulk structures and 0.15 and 0.30 for the surface structures (over the entire temperature range).

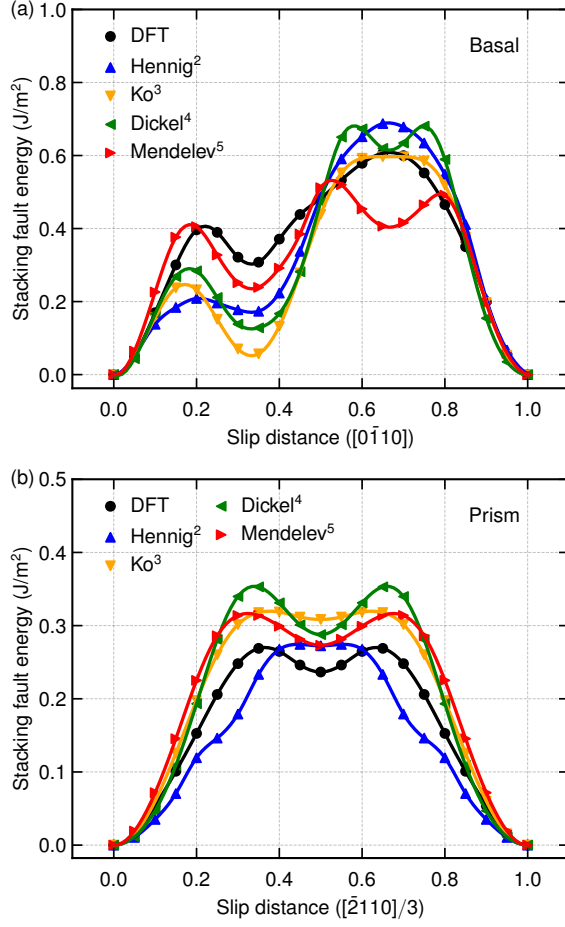
In each iteration in the bulk (surface) structure exploration, if the number of candidate configurations is larger than 100 (50) from the MD trajectory starting from the perturbed bulk (surface) supercells, 100 (50) randomly selected configurations are sent for DFT calculations (up to $100 \times 3 = 300$ ($50 \times 10 = 500$) configurations are sent for DFT calculations in one iteration).

* wang_han@iapcm.ac.cn

† zhaoxu@cityu.edu.hk

II. SUPPLEMENTARY FIGURES

Γ -lines from Different Empirical Potentials

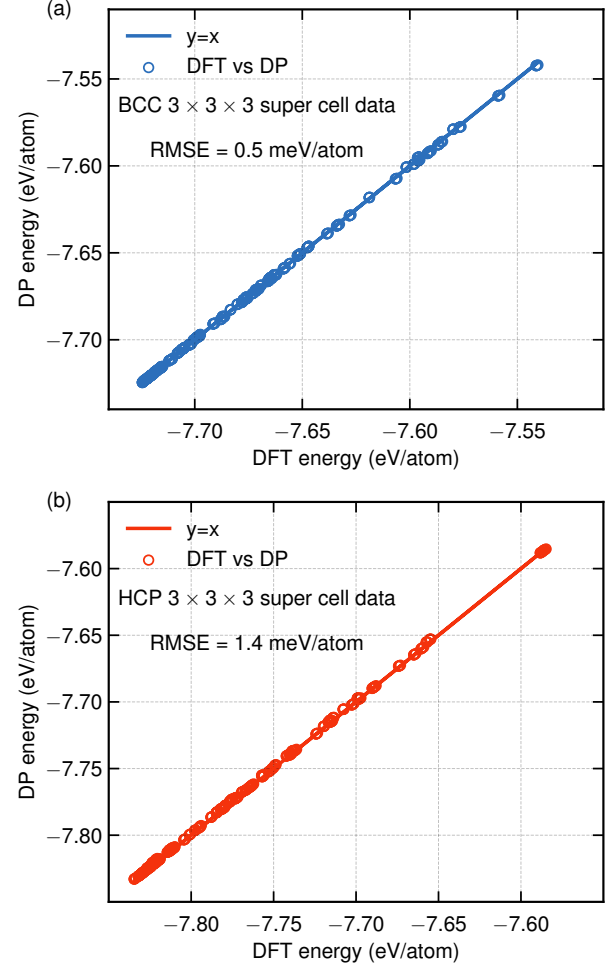


Supplementary Figure 1. The generalised stacking fault energy (γ -lines) on the (a) basal and (b) prism planes of HCP Ti calculated using DFT and empirical potentials (Hennig², Ko³, Dickel⁴, and Mendelev⁵).

Performance of DP for Larger Cell

We generated larger supercells ($3 \times 3 \times 3$ unit cells, 54 atoms; approximately 3.5X larger than original) for the BCC and HCP structures, calculated their properties and added them to the DP training datasets. This both enlarges the supercells and training set. (We did not study large supercells for the FCC structure, since it is considerably less important for Ti). The $3 \times 3 \times 3$ supercell datasets were constructed as follows. The equilibrium BCC and HCP supercells were generated and then affinely-scaled by 0.96, 0.98, 1.00, 1.02 and 1.04 the equilibrium lattice parameters. Random perturbations were applied to the atomic coordinates and supercell vectors, as described in Supplementary Methods (20 separate

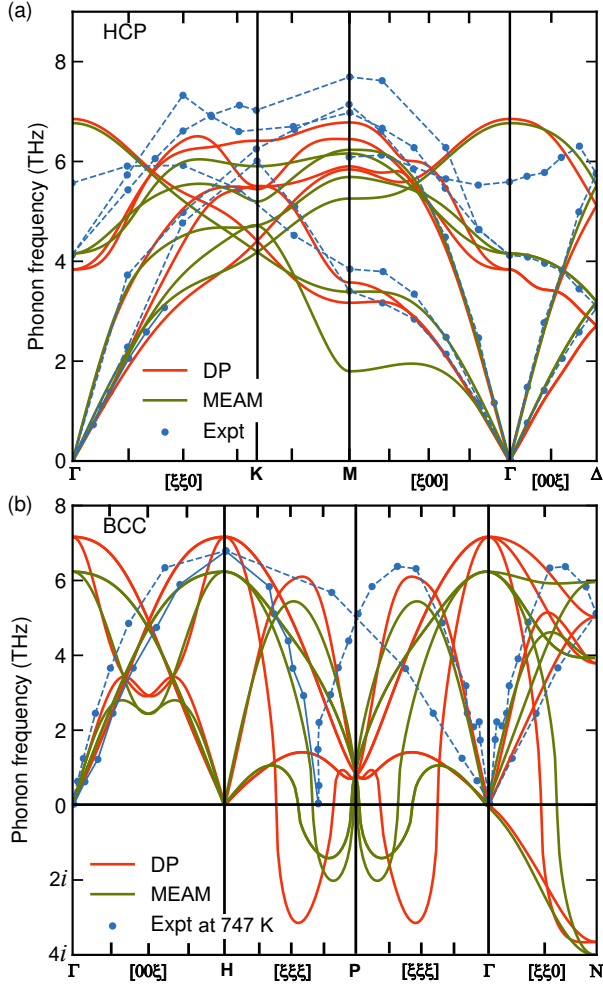
random perturbations were examined). *Ab initio* molecular dynamics (AIMD) simulations were also performed at 100 K using the same DFT parameters as that of the other DFT calculations (see Methods in the manuscript). After AIMD, 500 additional structure datasets were included for both the BCC and HCP phases. We use these datasets to test our DP and the results of the energy difference can be found in Supplementary Figure 2.



Supplementary Figure 2. DFT vs DP energies for $3 \times 3 \times 3$ (a) BCC and (b) HCP super cell data. The corresponding root mean square errors (RMSEs) are shown for each system.

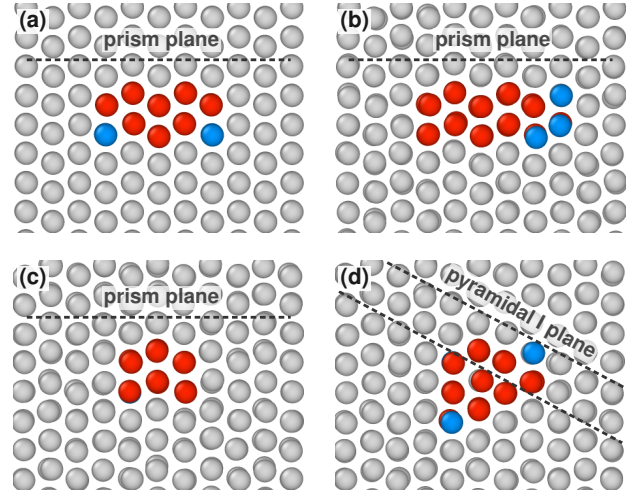
Phonon Spectra

The phonon spectra (Supplementary Figure 3) was calculated using the supercell approach. The force constants were first calculated using the $8 \times 8 \times 8$ supercell for both HCP and BCC Ti by LAMMPS. Then the phonon spectra was calculated using PHONOPY code⁶.



Supplementary Figure 3. The phonon spectra of (a) HCP and (b) BCC Ti from DP, MEAM², and experiment².

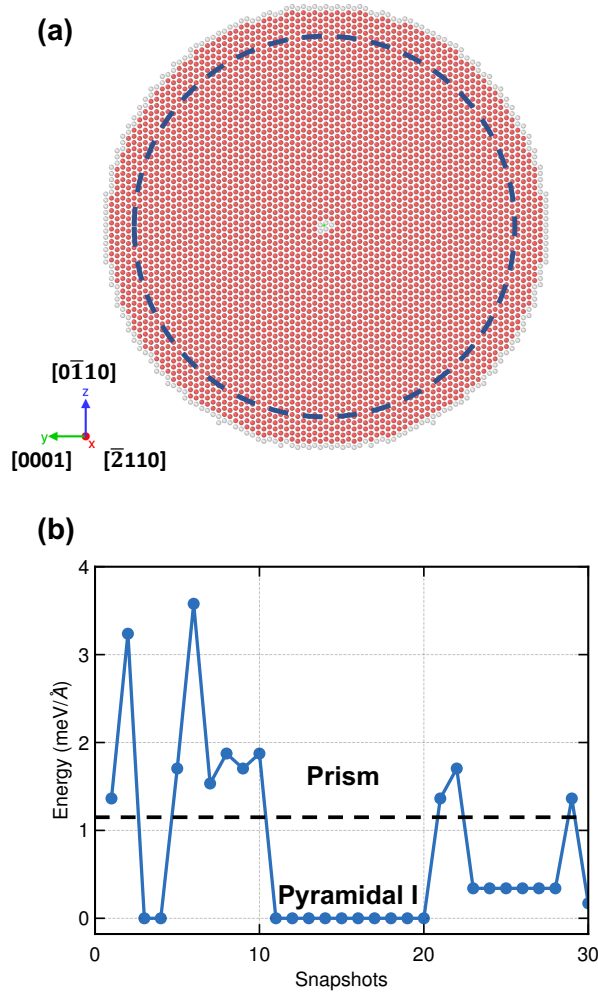
MD Simulations of the Screw $\langle a \rangle$ Dislocation



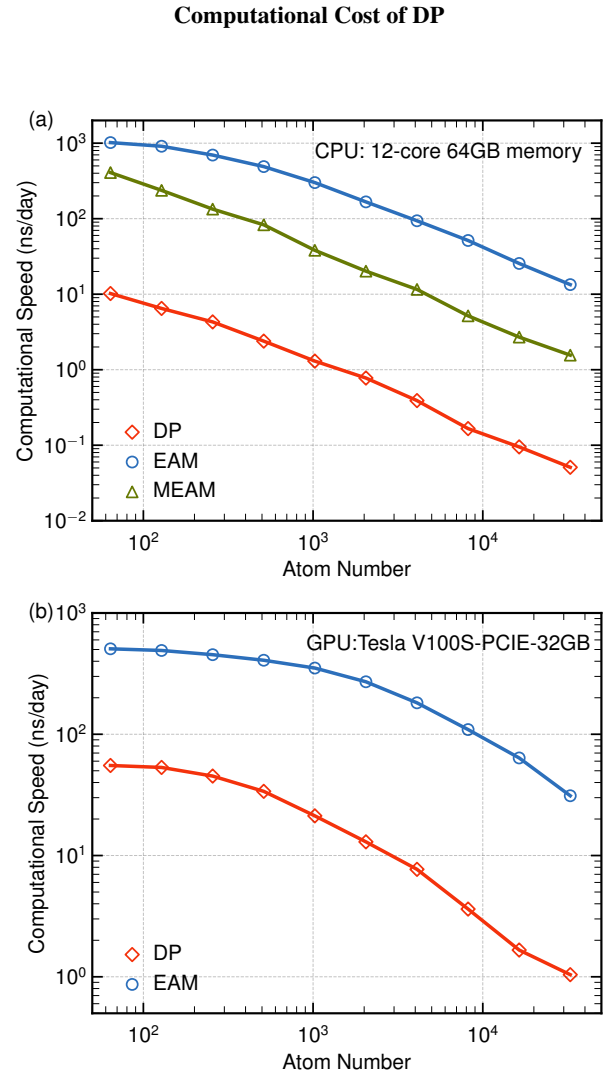
Supplementary Figure 4. MD simulation of the screw $\langle a \rangle$ dislocation on the prism plane at 300 K. (a) and (b) show dislocation dissociation on the prism plane, (c) is a compact core on the prism plane, and (d) shows a dislocation core with some spread along the pyramidal I plane.

Estimating the Dislocation Core Energy Difference

The cylinder dislocation configuration in Supplementary Figure 5(a) is used to estimate the dislocation core energy difference on different planes. A full screw $\langle a \rangle$ dislocation is initially inserted and the length of the dislocation line is $2a$ (~ 5.9 Å) with the radius of the cylinder ~ 92 Å. The atoms inside the blue dashed circle (radius ~ 80 Å) are allowed to move and the atoms outside are fixed according to the anisotropic elastic solution. The cylinder configuration is first relaxed and then MD simulations were performed at 600 K followed by energy minimisation of configurations at every 1 ps. This strategy is used to obtain dislocation core structures on both prism and pyramidal I planes. From Supplementary Figure 5(b), there are obviously two types of dislocation cores (Figs. 8(a) and (b) in the manuscript); the core energy of the prism dislocation is ~ 1.4 meV/Å higher than that of the pyramidal I dislocation.



Supplementary Figure 5. (a) Cylinder screw $\langle a \rangle$ dislocation configuration. (b) The total energy of different dislocation snapshots after minimisation. The prism dislocation core structure has higher energy.



Supplementary Figure 6. Speed comparison of the DP model, an EAM⁵, and/or an MEAM potential² on (a) CPU and (b) GPU. The time step is 1 fs. EAM on GPU uses single precision and DP on GPU uses double precision. The computational speed (ns/day) is read directly from the log file of LAMMPS.

III. SUPPLEMENTARY TABLES

Convergence Test of Vacancy Formation Energy

We calculated the vacancy formation energy E_v in the HCP structure as a function of supercell size (16 – 2000 atoms) using DP, EAM⁵, and MEAM² and up to 54 atoms in DFT; these data are presented in Supplementary Table I. These data shows E_v convergence to within 0.01 eV is achieved with 432 atoms for DP, EAM, and MEAM. The discrepancy between the DFT data and the DP, EAM, and MEAM E_v is not associated with simulation cell size.

Supplementary Table I. The dependence of the vacancy formation energy (eV) of HCP Ti on the supercell size for DFT, DP, EAM⁵, and MEAM².

Atom Number (Supercell)	DFT	DP	EAM	MEAM
16 ($2 \times 2 \times 2$)	2.03	1.97	1.59	1.98
24 ($2 \times 2 \times 3$)	2.01	1.99	1.61	2.02
36 ($3 \times 3 \times 2$)	2.07	2.45	1.69	2.08
54 ($3 \times 3 \times 3$)	2.06	2.41	1.70	2.18
128 ($4 \times 4 \times 4$)	-	2.45	1.73	2.18
432 ($6 \times 6 \times 6$)	-	2.41	1.73	2.18
1024 ($8 \times 8 \times 8$)	-	2.41	1.73	2.19
2000 ($10 \times 10 \times 10$)	-	2.41	1.73	2.19

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1. Plimpton, S. Fast parallel algorithms for short-range molecular dynamics. *J. Comput. Phys.* **117**, 1–19 (1995).
 2. Hennig, R., Lenosky, T., Trinkle, D., Rudin, S. & Wilkins, J. W. Classical potential describes martensitic phase transformations between the α , β , and ω titanium phases. *Phys. Rev. B* **78**, 054121 (2008).
 3. Ko, W.-S., Grabowski, B. & Neugebauer, J. Development and application of a Ni-Ti interatomic potential with high predictive accuracy of the martensitic phase transition. *Phys. Rev. B* **92**, 134107 (2015).
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