Supporting Information for

Point Defects at Grain Boundaries Can Create Structural Instabilities and Persistent Deep Traps in Metal Halide Perovskites

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Details of Training of Machine Learning Force Field

We employed the mean-squared error of energy and forces as our loss function, as shown below.

$$L_E(x;\theta) = \frac{1}{N}\left(E(x;\theta) - E^*\right)^2$$

$$L_F(x;\theta) = \frac{1}{3N}\sum_{k=1}^{N}\sum_{\alpha=1}^{3}\left(F_{k,\alpha}(x;\theta) - F_{k,\alpha}^*\right)^2$$

where α represents the α-th component of the force on atom k, and the superscript * indicates the reference value of the property that should be provided beforehand. Averaging each loss over atomic contributions before applying weights ensures that each property's loss is fairly represented in the total loss

$$L = p_e L_e + p_f L_f$$

Here, $L_e$and L_f denote the loss in energy and forces, respectively, and $p_e$and $p_f$give the prefactors of the energy and force losses. Figure S1 presents comparison of machine learning and ab initio potential energies. Figure S2 shows evolutions of the learning curves of energy and force losses.

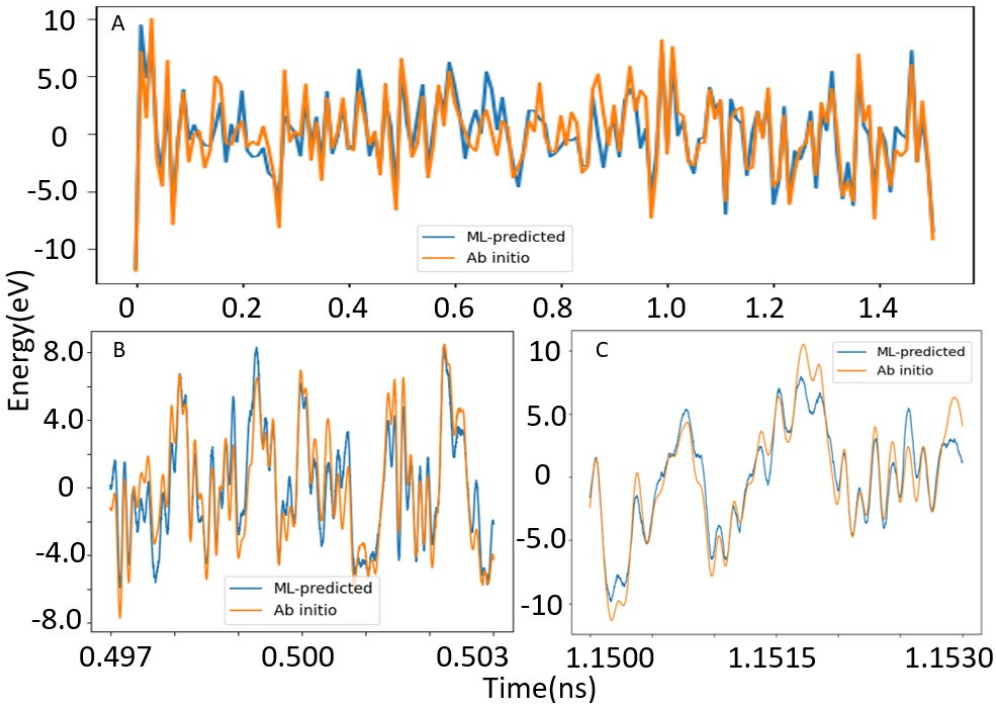

Figure S1. Comparison of machine learning and ab initio potential energies for **(A)** full 1.5 ns trajectory, **(B)** and **(C)** selected trajectory parts. The ab initio calculations are performed every 10 ps for the full trajectory, and every 2 fs for the selected parts. The root means square error of the machine learning potential energy relative to the ab initio potential energy for the whole trajectory is 6.58 meV per atom, within recommended machine learning force field accuracy, especially for such a semi-amorphous system.

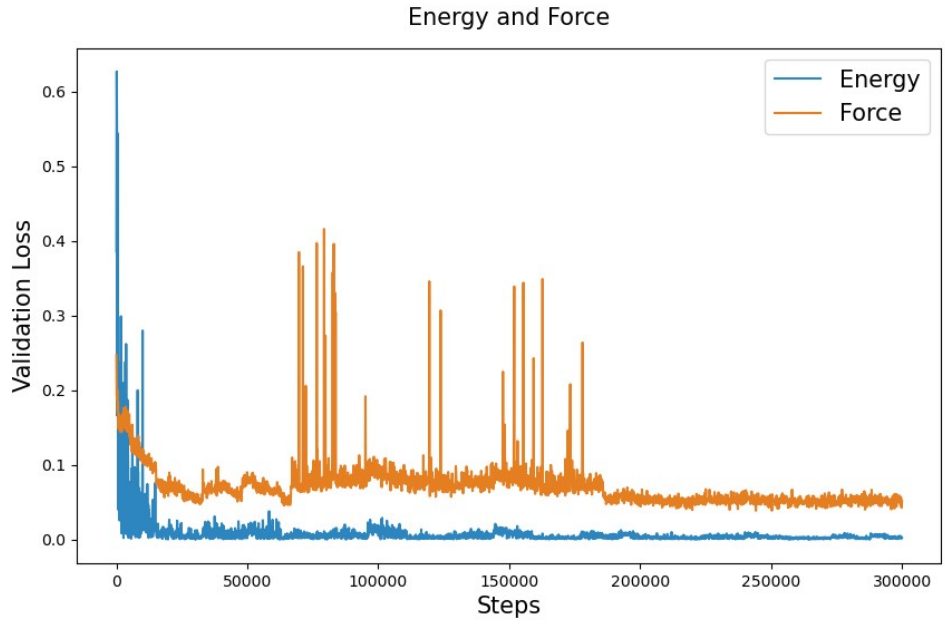

Figure S2. Evolution of the learning curves of energy and forces for the validation set.

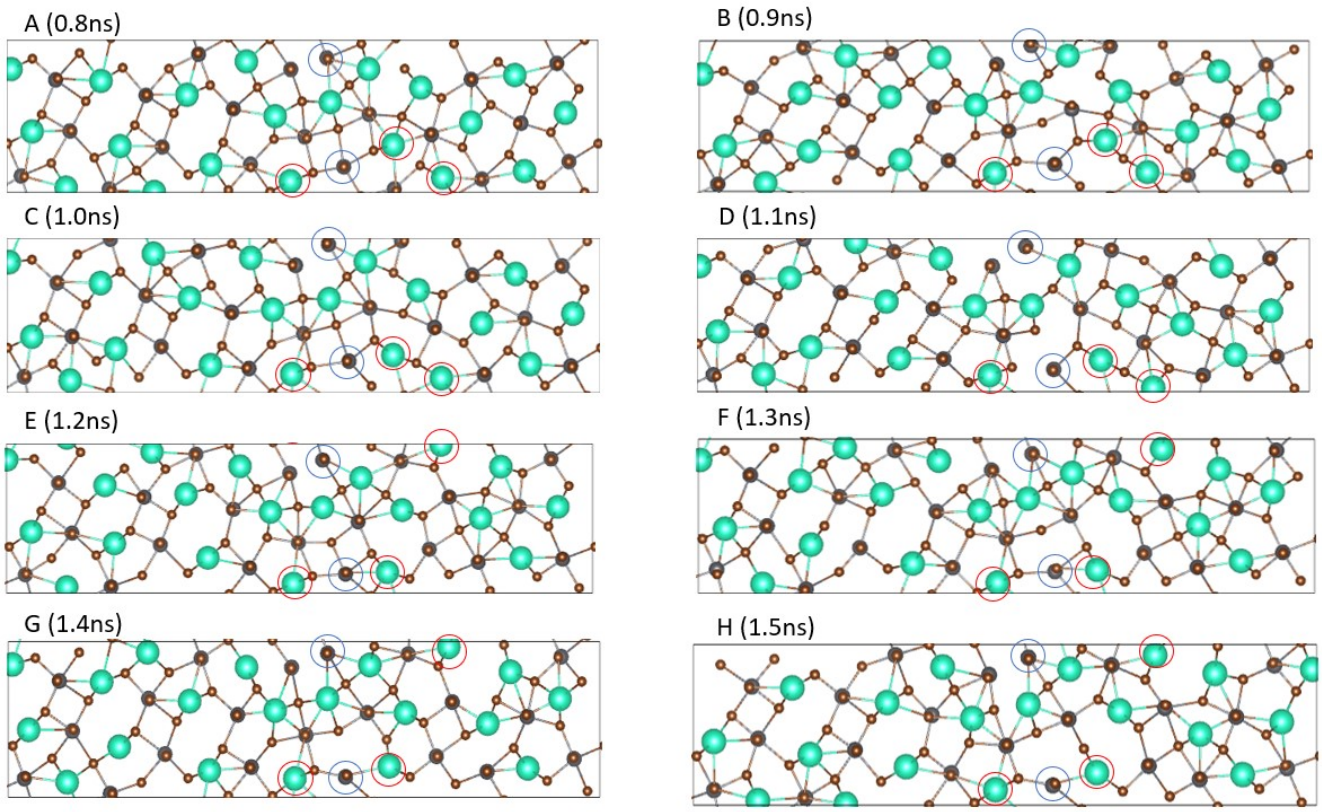

Figure S3. (A-H) atomic configurations at different times showing the grain boundary slide around 1.2 ns, as depicted schematically in Figure 2A. The Pb atoms used to report the Pb-Pb distance in Figure 2C are circled in blue. The Cs atoms whose average position is shown in Figure 2D are circled in red. Note that the Cs atoms slide to the bottom and appear on the top of the simulation cell due to the periodic boundary conditions.

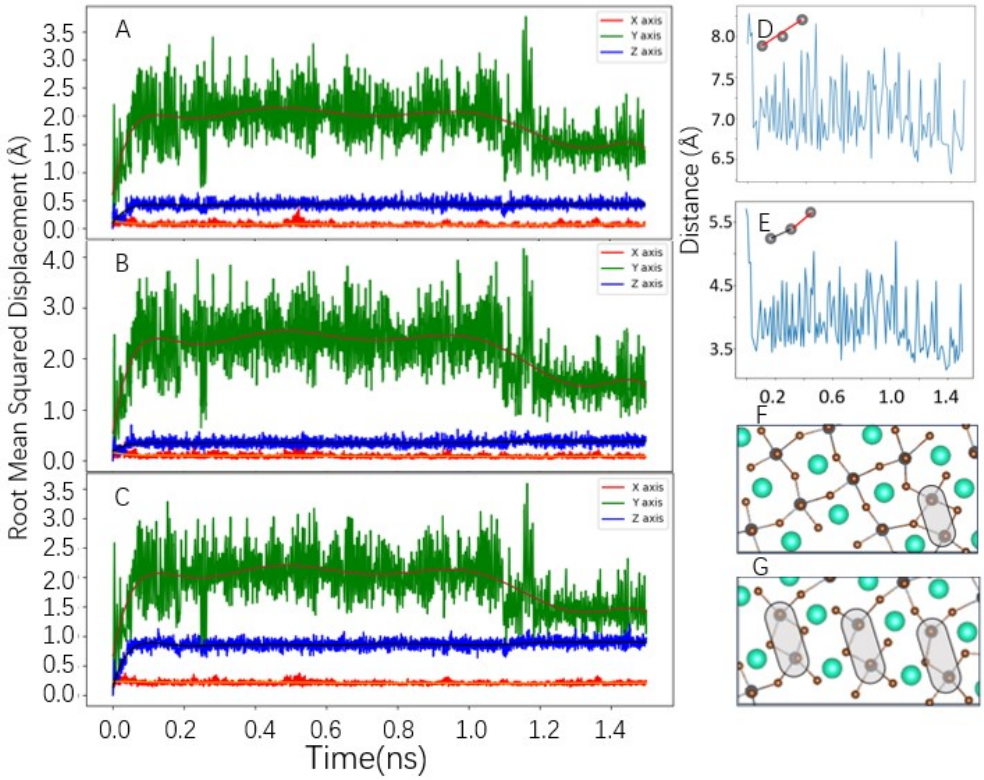

Figure S4. Mean squared displacement (MSD) of **(A)** Cs, **(B)** Pb, and **(C)** Br atoms in the X, Y and Z directions of the simulation cell along the 1.5 ns trajectory. **(D-E)** Evolution of the distances between the Pb atoms in the Pb trimer along the 1.5 ns trajectory. The considered distance is indicated by the red line in the trimer picture in the inserts. **(F–G)** Screenshots of Pb pairs in the sub-boundary region before and after the atomic rearrangement at 20 ps and 40 ps, respectively. The evolution curves shown in parts A-E identify the initial GB relaxation event at the early time (~30 ps), and the subsequent GB sliding and tilting at around 1.1 ns. Parts F-G demonstrate formation of additional Pb pairs after the initial GB relaxation.

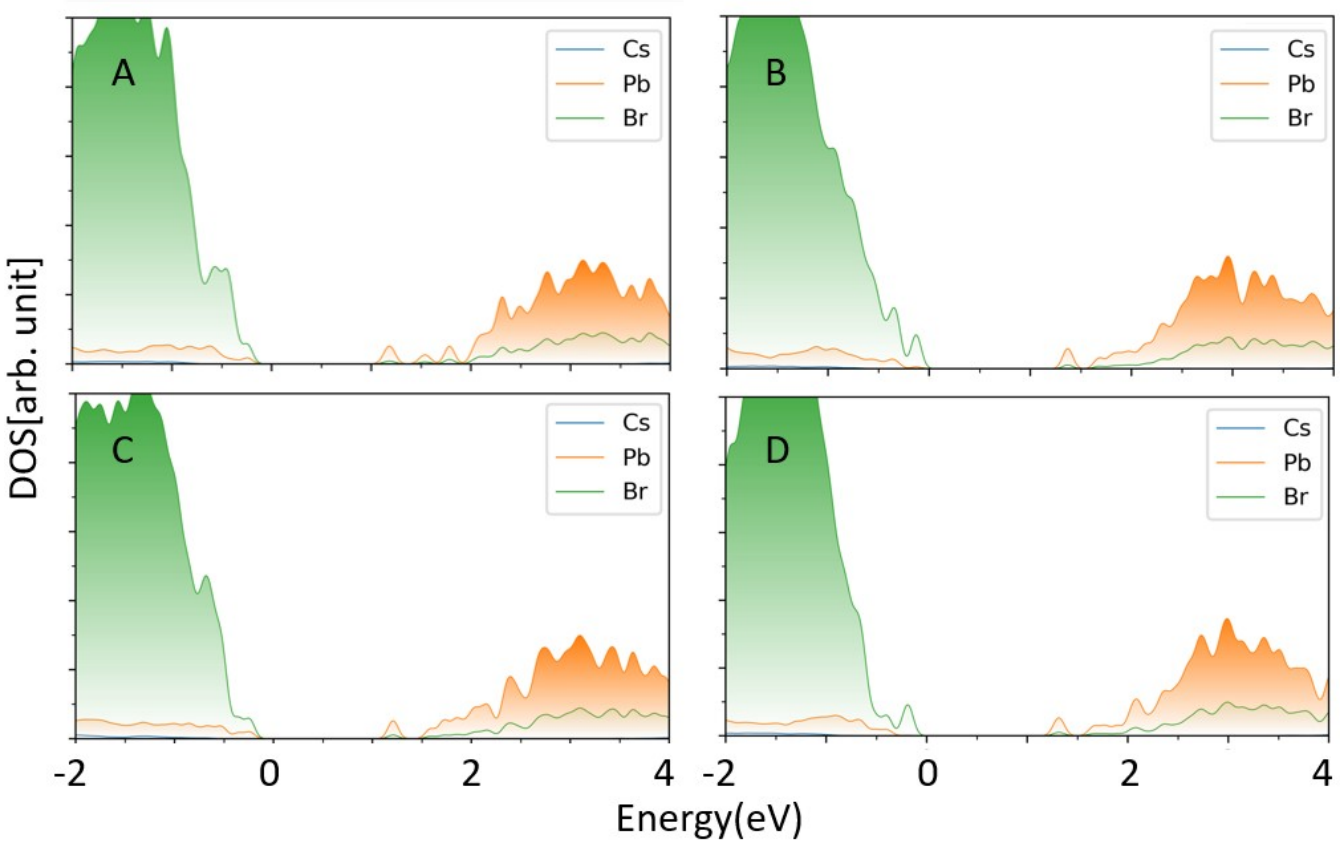

Figure S5. Representative densities of states (DOS) at different times in the middle of the 1.5 ns trajectory: **(A)** 498,557 fs, **(B)** 499,705 fs, **(C)** 500,160 fs, and (**D)** 500,500 fs. The DOS shows a trap state that comes from a Pb-Pb-Pb trimer. In comparison, the DOS of the optimized structure, Figure 1B, shows two electron traps. The shallower trap can disappear, because the grain boundary is sufficiently adaptable to allow the defect to heal.

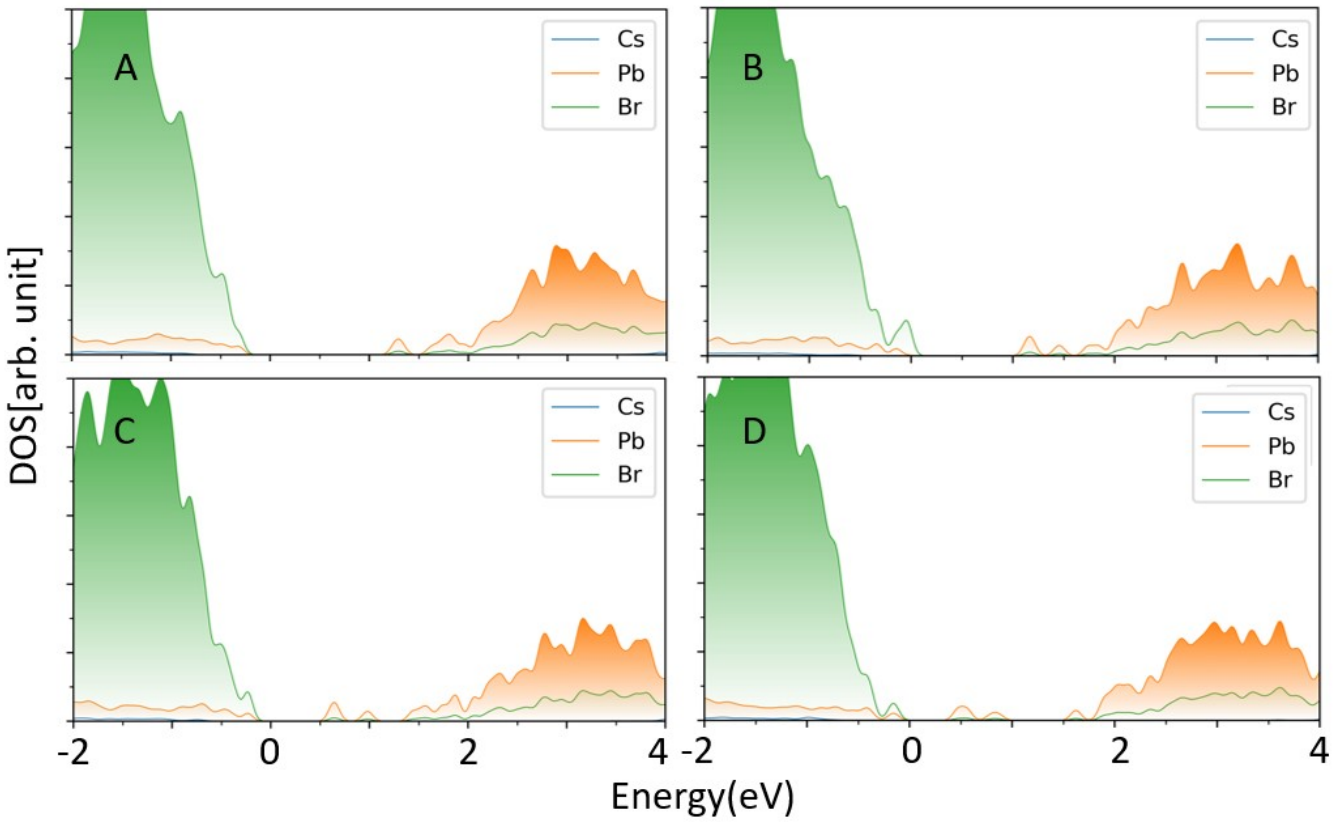

Figure S6. Representative densities of states (DOS) at different times after the grain boundary starts sliding, Figure 2B: **(A)** 1,150,435 fs, **(B)** 1,150,921fs, **(C)** 1,151,151 fs, and (**D)** 1,152,050 fs. The DOS shows a deep trap state that comes from a Pb-Pb-Pb trimer, as well as other trap states that appear during the sliding stage.