Supporting Information for

Local Order and Rotational Dynamics in Mixed A-Cation Lead Iodide Perovskites

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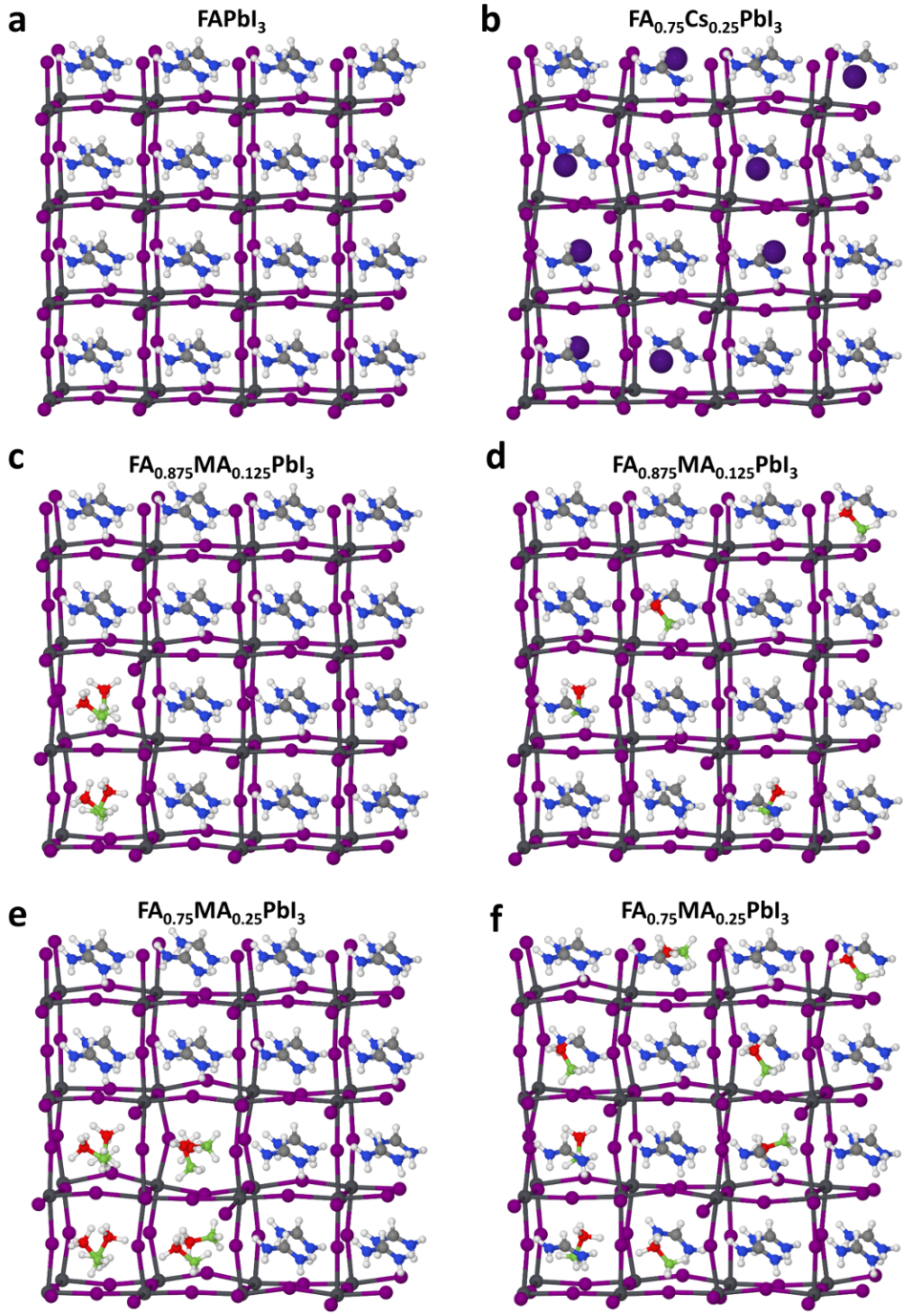

Figure S1: Initial configurations used for the molecular dynamics calculations. (a) $FAPbI_3$, (b) $FA_{0.75}Cs_{0.25}PbI_3$, (c) $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions clustered in the corner of the computational box (d) $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box, (e) $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions clustered in the corner of the computational box (f) $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box. Periodic boundary conditions are applied in all cases. The C and N atoms of MA^+ and FA^+ have different colors for clarity.

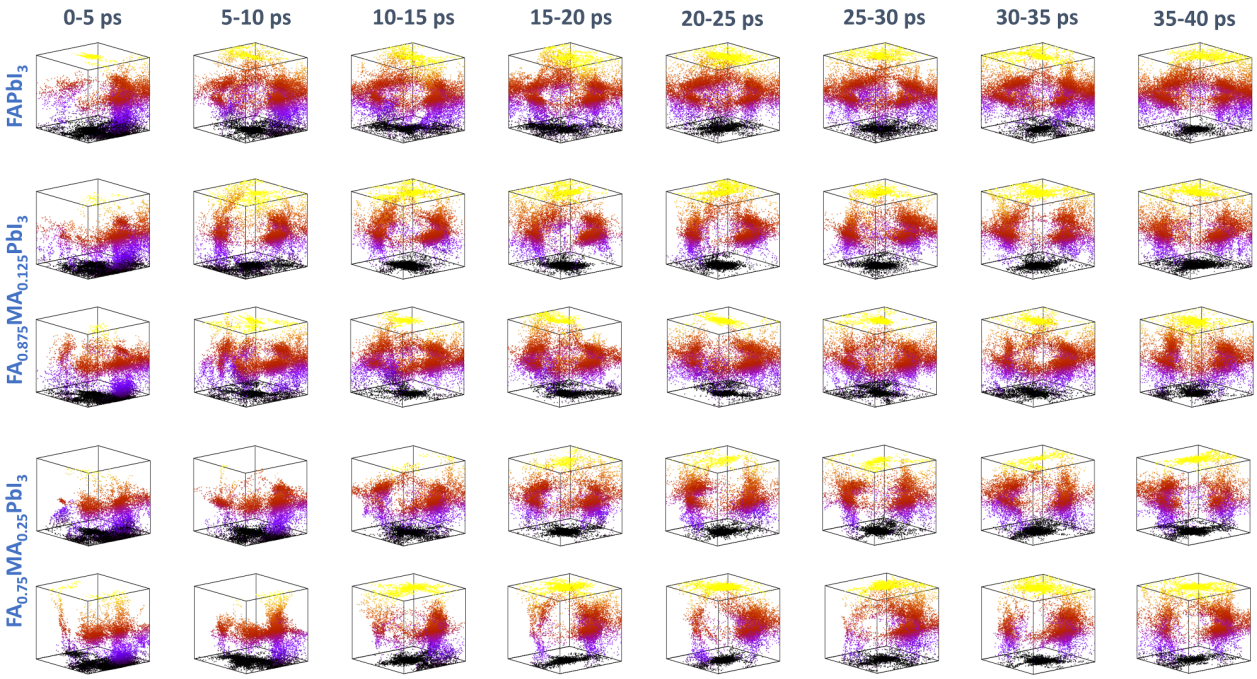

Figure S2: Orientations of the molecular dipoles of the FA^+ cations projected on the cubic inorganic cage for every 5 ps of simulation time. From upper to lower: $FAPbI_3$, $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box, $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions clustered in the corner of the computational box, $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box, $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions clustered in the corner of the computational box.

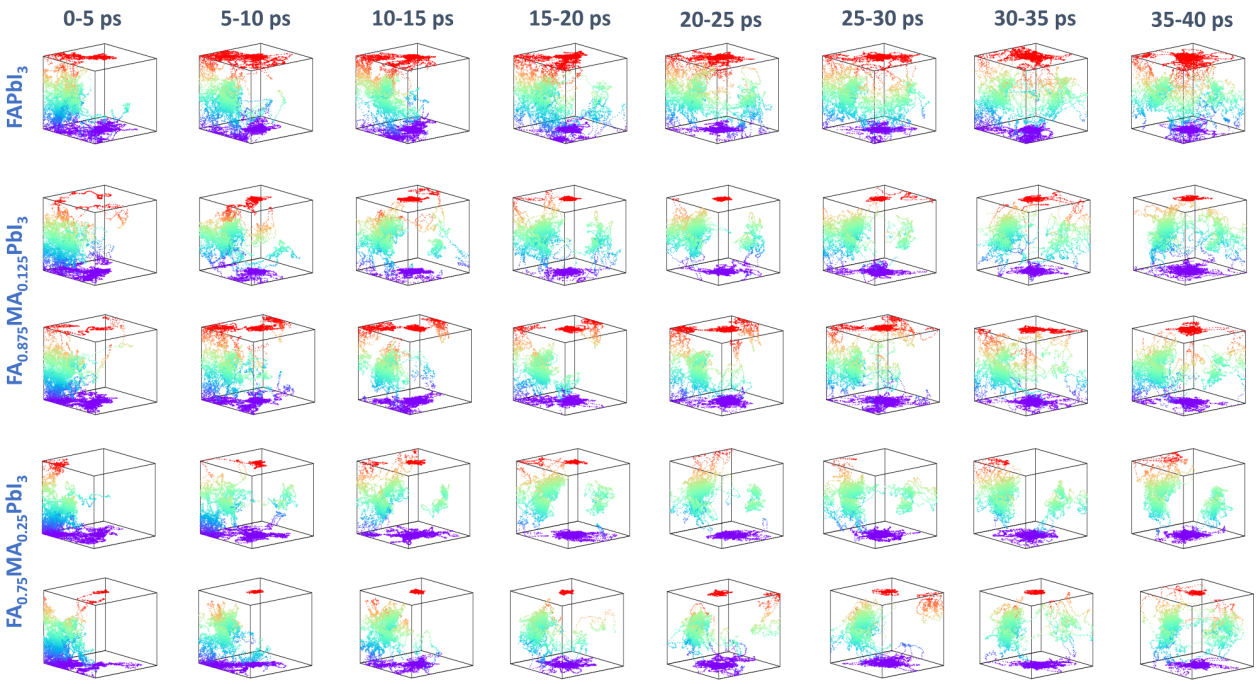

Figure S3: Orientations of the N-N axes of the FA^+ cations projected on the cubic inorganic cage for every 5 ps of simulation time. From upper to lower: $FAPbI_3$, $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box, $FA_{0.875}MA_{0.125}PbI_3$ with the MA^+ ions clustered in the corner of the computational box, $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions homogeneously distributed within the computational box, $FA_{0.75}MA_{0.25}PbI_3$ with the MA^+ ions clustered in the corner of the computational box.

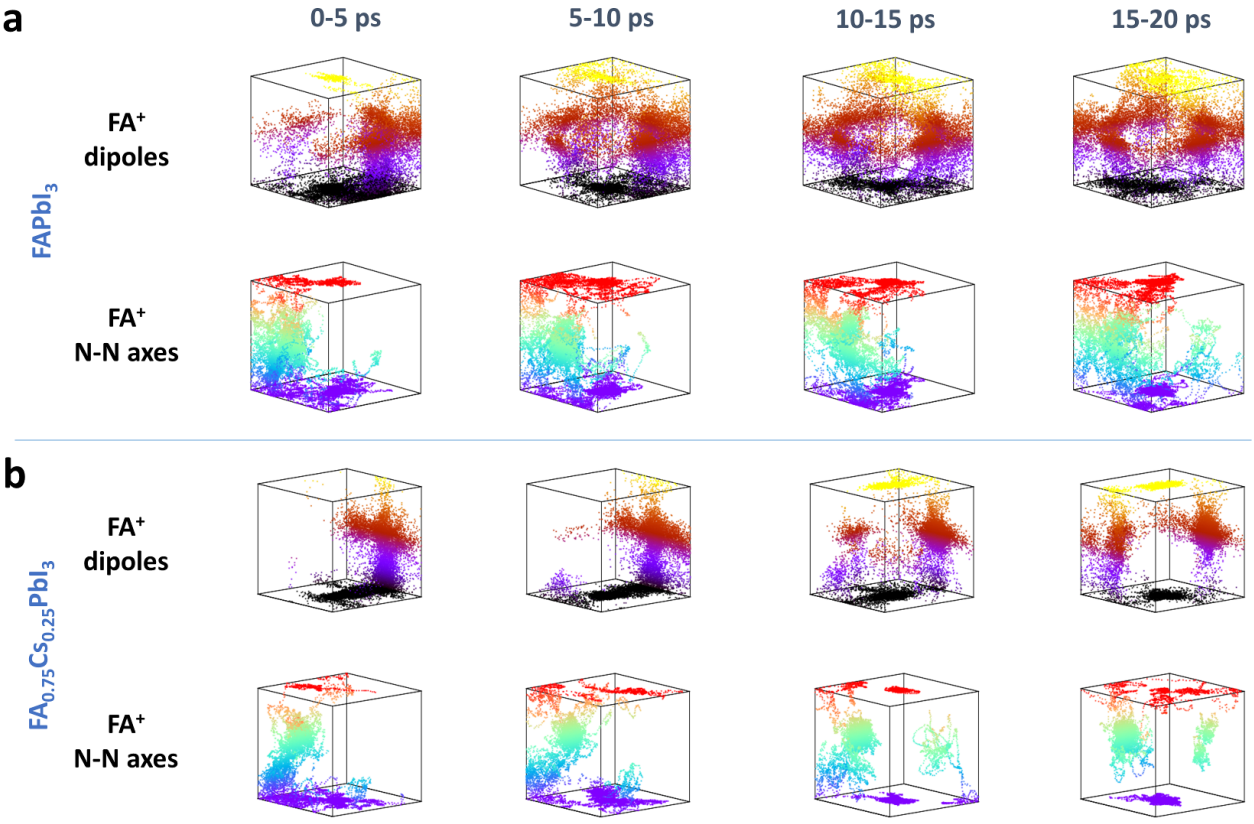

Figure S4: Orientations of the molecular dipoles (upper) and the N-N axes (lower) of the FA^+ cations projected on the cubic inorganic cage for every 5 ps of simulation time in the case of (a) $FAPbI_3$ and (b) $FA_{0.75}Cs_{0.25}PbI_3$.

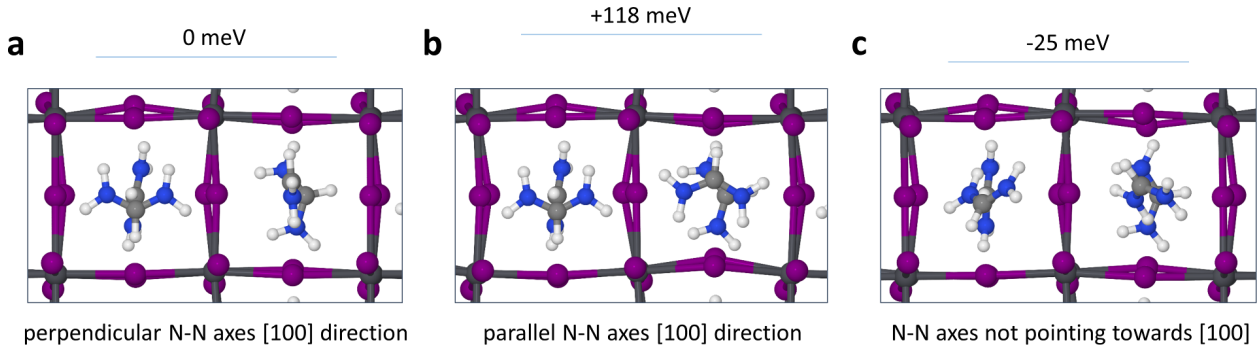

Figure S5: Comparison of the ground state energy for a $FAPbI_3$ system in a $2 \times 2 \times 2$ supercell having two of the FA^+ cations with (a) perpendicular N-N axes pointing towards $\langle 1\,0\,0 \rangle$ directions, (b) perpendicular N-N axes pointing towards the same $[1\,0\,0]$ direction and (c) N-N axes not pointing towards $\langle 1\,0\,0 \rangle$ directions.