MACE Foundation Models for Lattice Dynamics:

A Benchmark Study on Double Halide

Perovskites

Supporting Information

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Database Overview

Data availability

All DFT data for our HDP database is stored in the Harvard Dataverse, which can be freely accessed from the following links:

1. Fluoride HDPs: https://doi.org/10.7910/DVN/WBOXPG

2. Chloride HDPs: https://doi.org/10.7910/DVN/JGODBE

3. Bromide HDPs: https://doi.org/10.7910/DVN/RIMZ2F

4. Iodide HDPs: https://doi.org/10.7910/DVN/ATZEFE

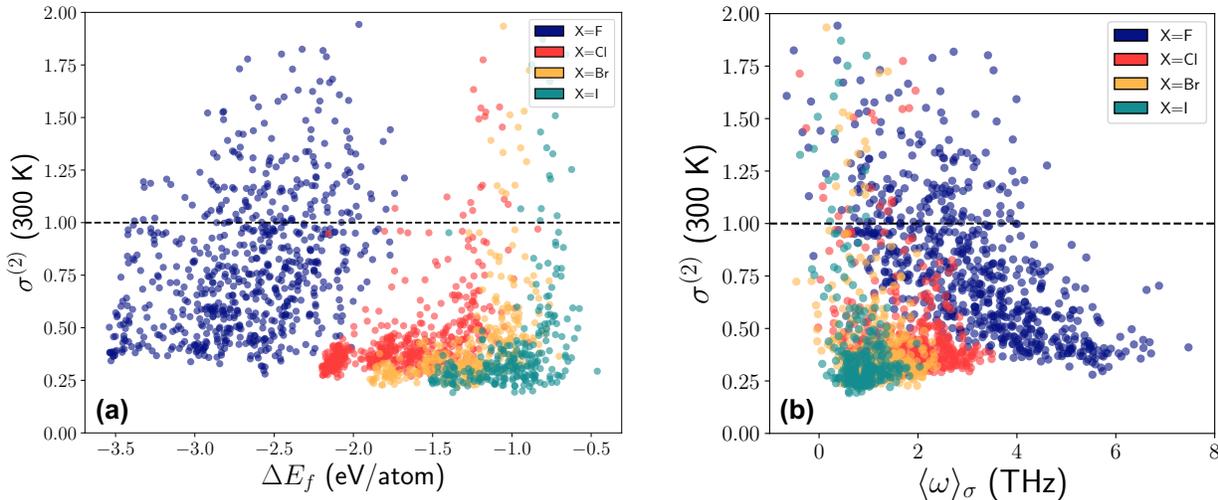## Vibrational anharmonicity landscape at DFT level of theory

Figure S1: Landscape of room-temperature vibrational anharmonicities as measured by $\sigma^{(2)}$ as a function of formation energies for HDPs, in which data results for HDPs with different halogen anions are separately colour-coded. (a) $\sigma^{(2)}$ plotted as a function of the formation energies ΔE_f. (b) $\sigma^{(2)}$ plotted as a function of anharmonicity-weighted-averaged phonon frequency for each HDP, defined as $\langle \omega \rangle_\sigma = \sum_{\boldsymbol{q},n} \omega(\boldsymbol{q},n)\sigma^{(2)}(\boldsymbol{q},n)/\sum_{\boldsymbol{q},n}\sigma^{(2)}(\boldsymbol{q},n)$, in which the phonon-mode-resolved anharmonicity score $\sigma^{(2)}(\boldsymbol{q},n)$ was computed using the same definition as Eq. (1) except all the atomic forces are projected onto individual phonon eigenvectors $\boldsymbol{u}(\boldsymbol{q},n)$ [Reproduced from Yang *et al.*[1]].

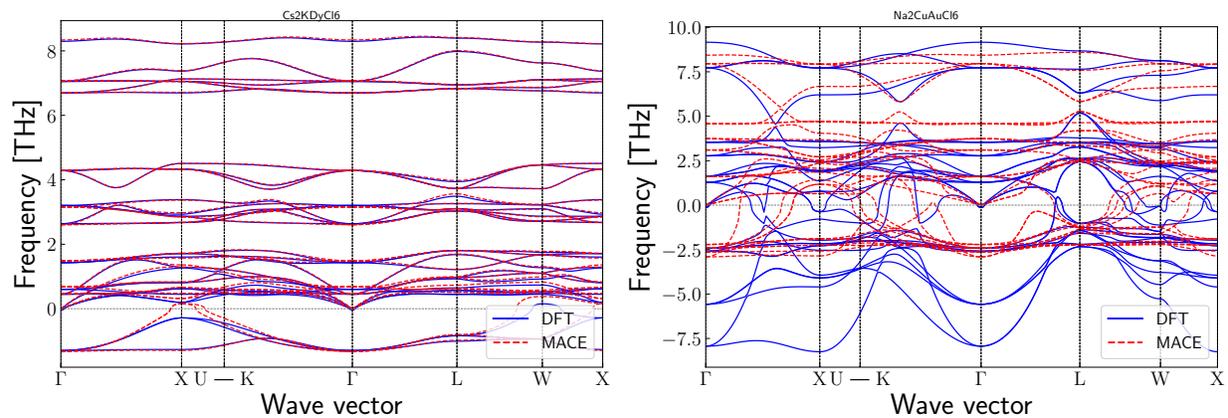# Exemplary Phonon Dispersion Curves

Figure S2: Comparisons of the phonon dispersion relationship computed with DFT and `omat-0-medium` foundation model. The results shown above correspond to the compounds of (Left) Cs_2KDyCl_6 and (Right) $Na_2CuAuCl_6$, which are the best and worst performing compounds for predicting harmonic phonon properties with the `omat-0-medium` model, respectively.

Accuracies of Predicting Harmonic Phonon Properties: Breakdown Analysis in Different Chemical Spaces
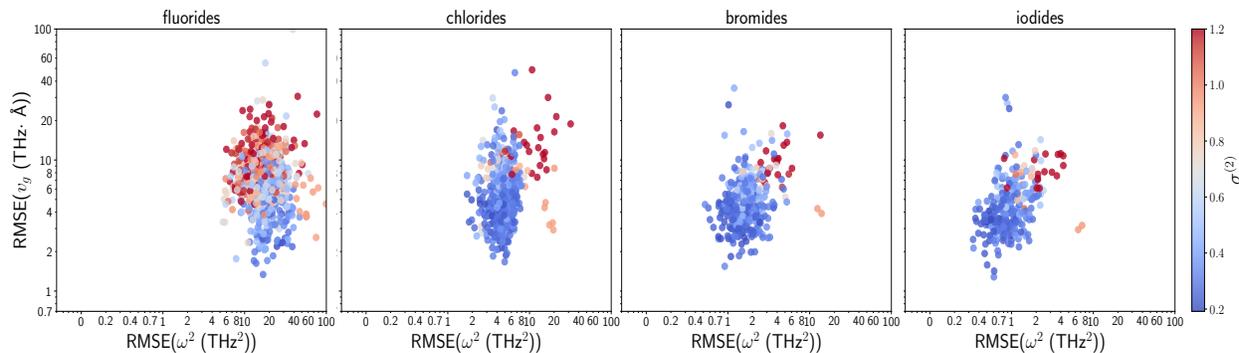

Figure S3: Scatter plots showing the correlations between the root-mean-squared-errors in predicting the phonon eigenfrequencies and group velocities using the MACE foundation model with respect to the DFT results. Results for HDPs with different halide anions are presented in separate subplots to better highlight the chemical trend. Each point in the plots are colour-coded according to their anharmonicity scores $\sigma^{(2)}$ obtained from DFT calculations.[1] For this set of results, the `matpbs-pbe-omat-ft` model was used.
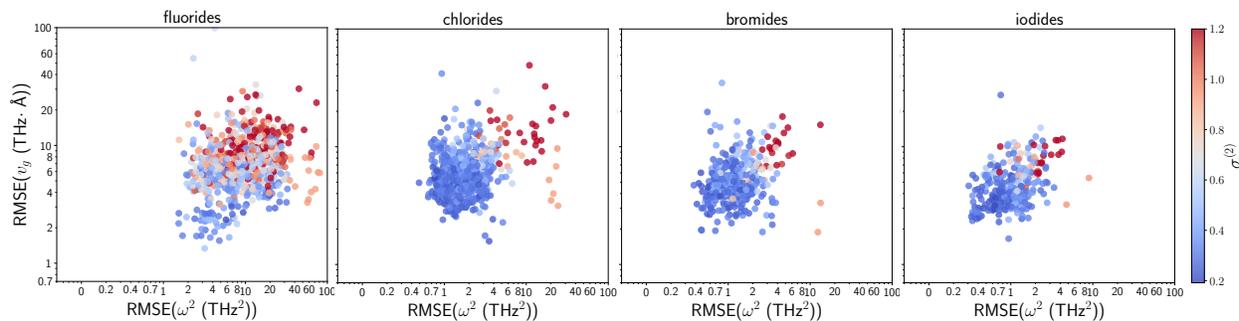

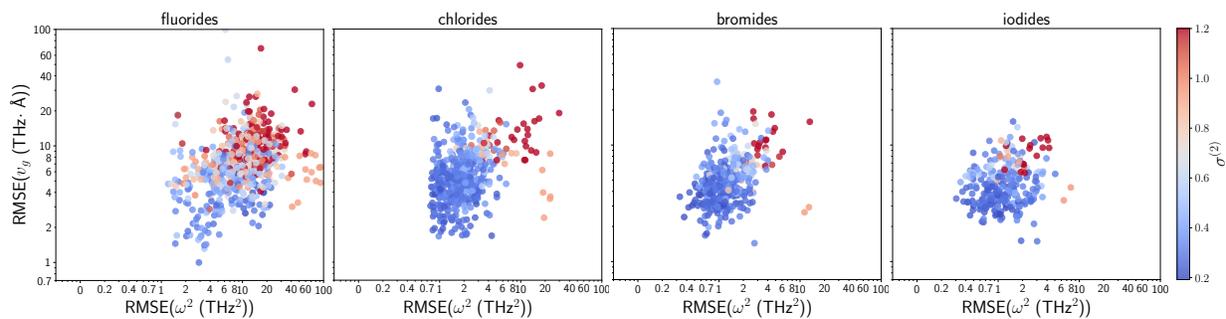Figure S4: Same as Fig. S3 with results obtained using the `mpa-0-medium` model.

Figure S5: Same as Fig. S3 with results obtained using the `mp-0b3-medium` model.
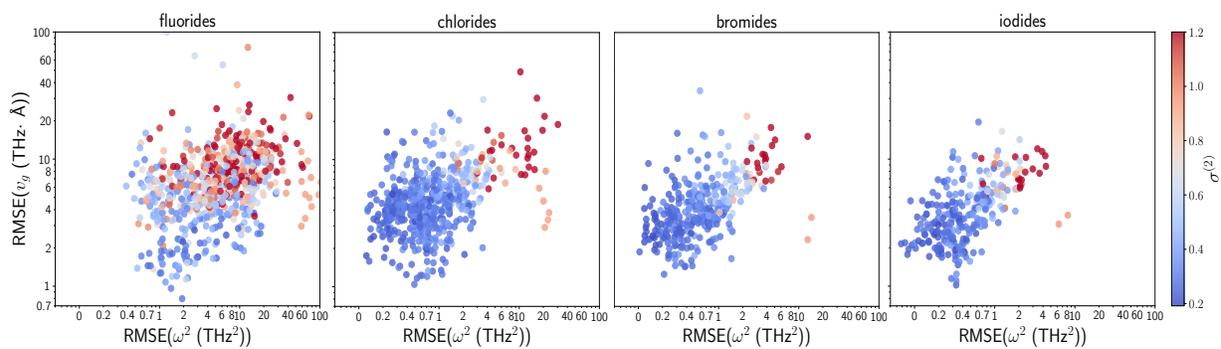

Figure S6: Same as Fig. S3 with results obtained using the `omat-0-medium` model.

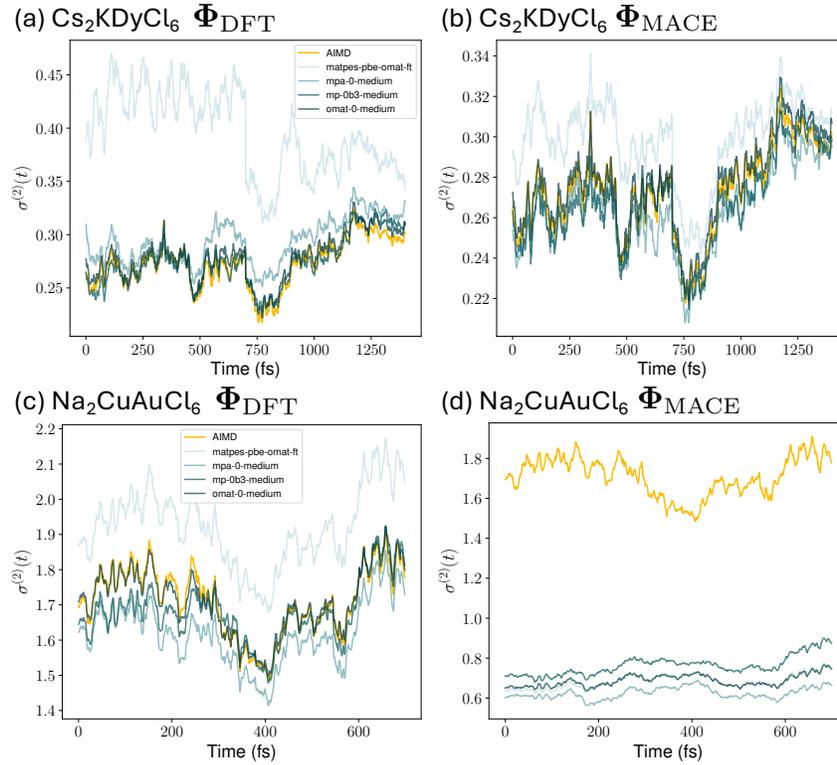# Accuracies of Predicting the Room-Temperature Vibrational Anharmonicity

Figure S7: Examples of the $\sigma^{(2)}(t)$ trajectories for two exemplary chloride HDPs [Fig. 1(a)]. The anharmonic scores $\sigma^{(2)}$ are determined for the configurations that were previously sampled from AIMD,[2] with the atomic forces for each trajectory frame recomputed by different MACE foundation models. On the left panel, we compare the results whereby the harmonic components of the atomic forces were determined based on DFT-derived force constants (Φ_{DFT}), whereas the right panel shows the case for Φ_{MACE}, in which the force constants were also recomputed using the corresponding MACE foundation models.
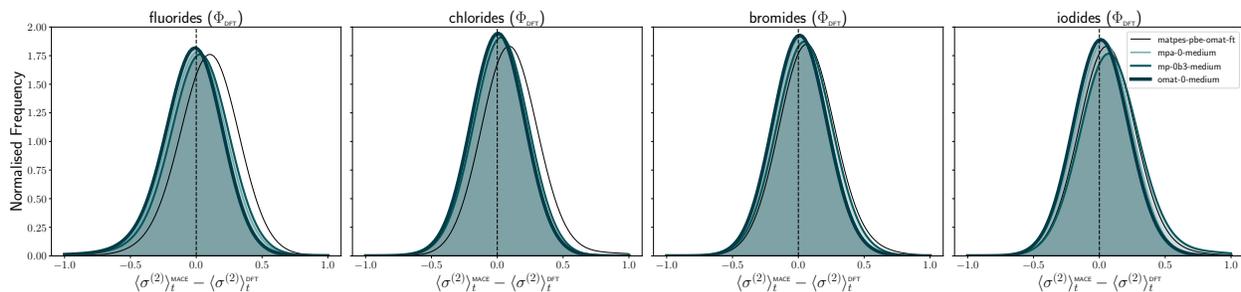
Figure S8: Statistical distributions on the differences in the trajectory-averaged anharmonic scores computed from MACE foundation models and DFT ($\langle\sigma^{(2)}\rangle_t^{\mathrm{MACE}} - \langle\sigma^{(2)}\rangle_t^{\mathrm{DFT}}$), both evaluated on the AIMD trajectories. The harmonic force constant matrix calculated from DFT (Φ_{DFT}) are used for determining the harmonic component of the atomic forces. Data for DHPs with different halide anions are shown separately.
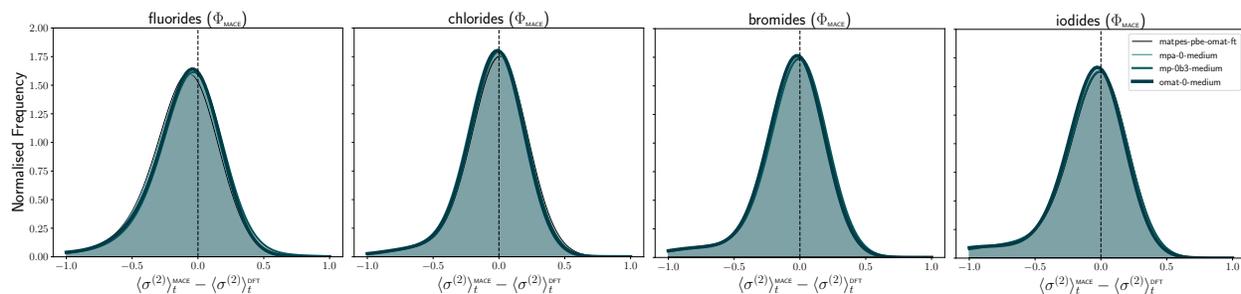

Figure S9: Same as Fig. S8, except the force constant matrix (Φ_{MACE}) are computed from the finite-difference approach using the same MACE foundation model for evaluating the total atomic forces on AIMD trajectory frames.
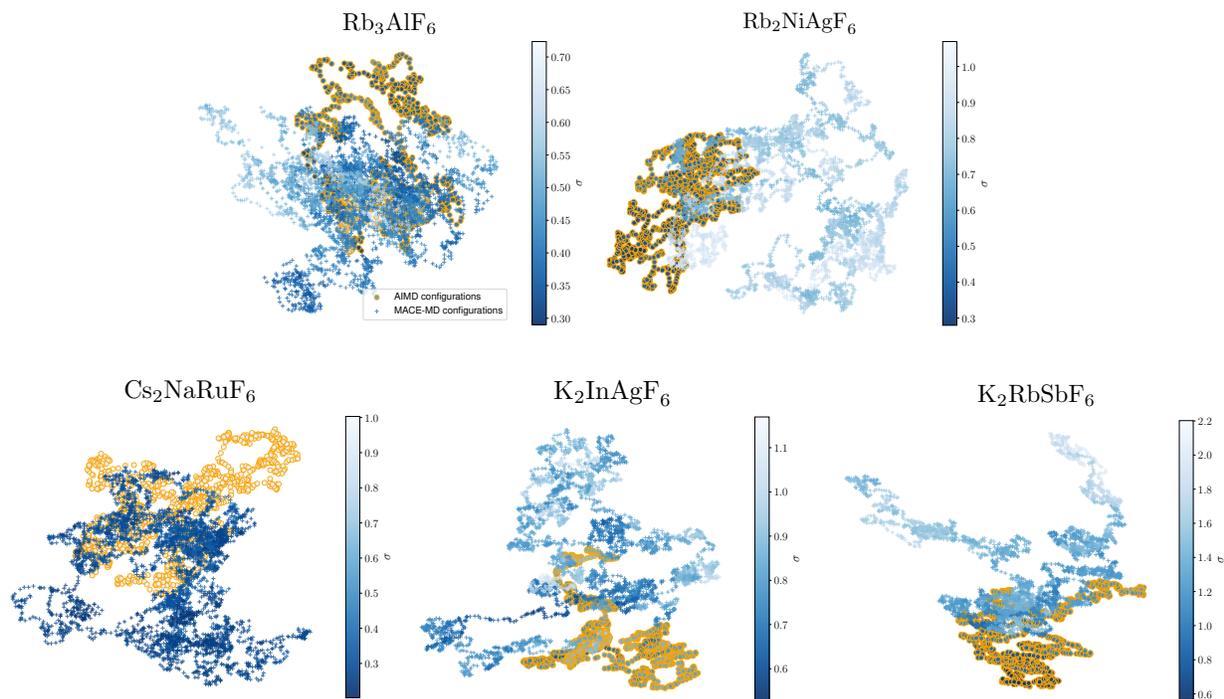
Additional Sketch Map Analysis

Figure S10: KPCA maps that compare the configurational space sampled by AIMD[1] and MACE-MD with the `omat-0-medium` model. Each configuration (point on the KPCA map) is further colour-coded with its anharmonicity score $\sigma^{(2)}$. The harmonic force components necessary for computing $\sigma^{(2)}$ were determined from the force constants that are computed with the same energy model as for the MD simulations.

Rb$_3$AlF$_6$

Rb$_2$NiAgF$_6$

Cs$_2$NaRuF$_6$
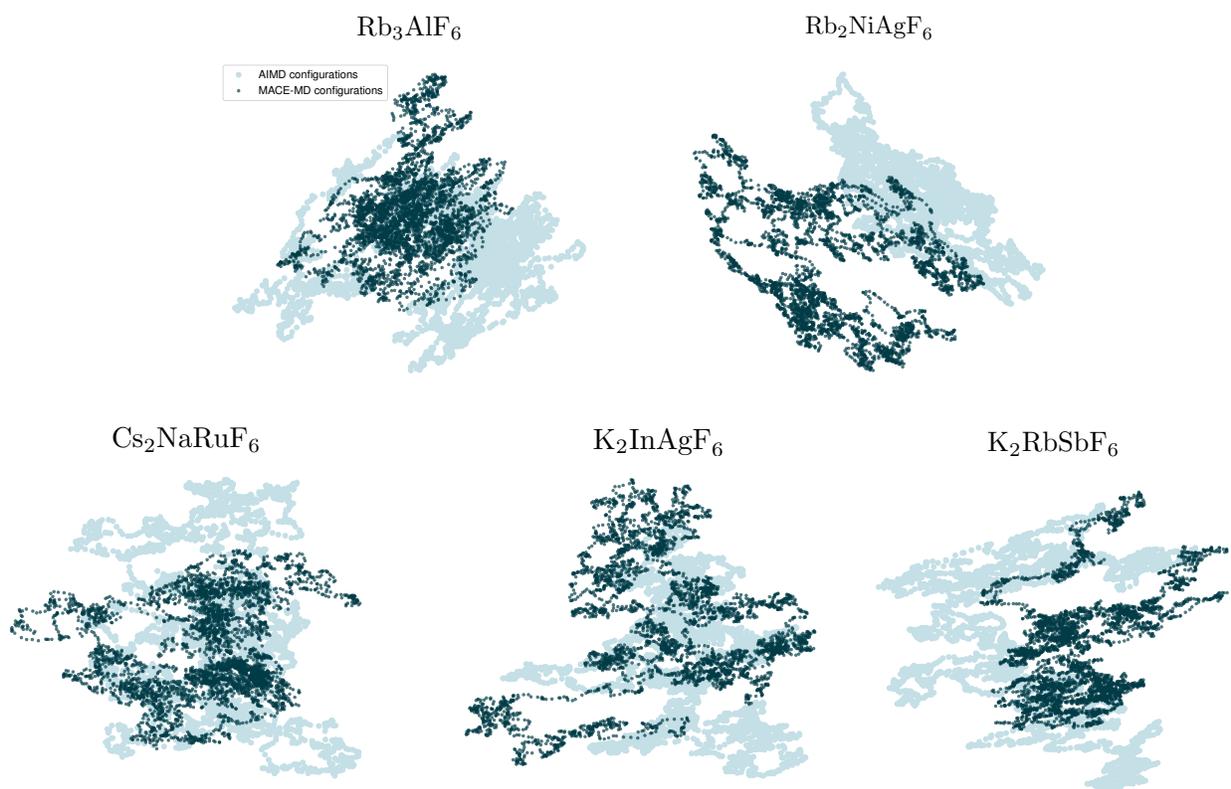
K$_2$InAgF$_6$

K$_2$RbSbF$_6$

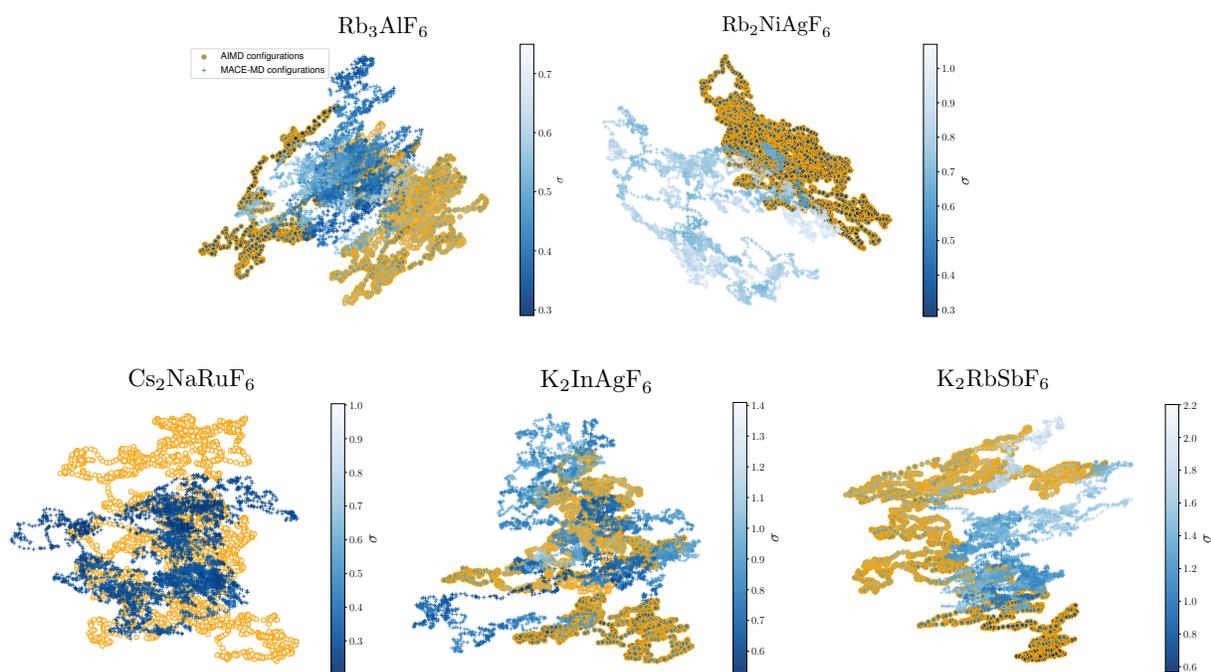Figure S11: Same as Fig. 5 which now includes a longer AIMD trajectory (4 ps in total) for each compound.

Figure S12: Same as Fig. 5 which now includes a longer AIMD trajectory (4 ps in total) for each compound.

References

(1) Yang, J.; Fan, J.; Li, S. Mapping the room-temperature dynamic stabilities of inorganic halide double perovskites. *Chem. Mater.* **2022**, *34*, 9072.

(2) Yang, J. Mapping temperature-dependent energy–structure–property relationships for solid solutions of inorganic halide perovskites. *J. Mater. Chem. C* **2020**, *8*, 16815.