Supplementary Information (SI) for Journal of Materials Chemistry C.

Machine learning potential-assisted design of thermoelectric performance in anharmonic $CsPbBr_3$ and $CsPbI_3$

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TABLE S1. DFT datasets preparation for machine learning. AIMD simulation based on PBEsol functional within the NVT ensemble is used to generate atomic configurations deviated from equilibrium. The simulation at specified temperature is performed with a time step of 1 fs. The atomic information including reference energy, force and virial tensor can be extracted from SCF with the convergence threshold of 10^{-6} eV/atom. A total of 900 configurations are extracted from AIMD simulation.

Provided structure	100K	200K	300K	400K	500K	600K	700K	800K	900K
Orthorhombic $CsPbBr_3$	√	√	√	√	√	√	√	√	√
Orthorhombic $CsPbI_3$	√	√	√	√	√	√	√	√	√

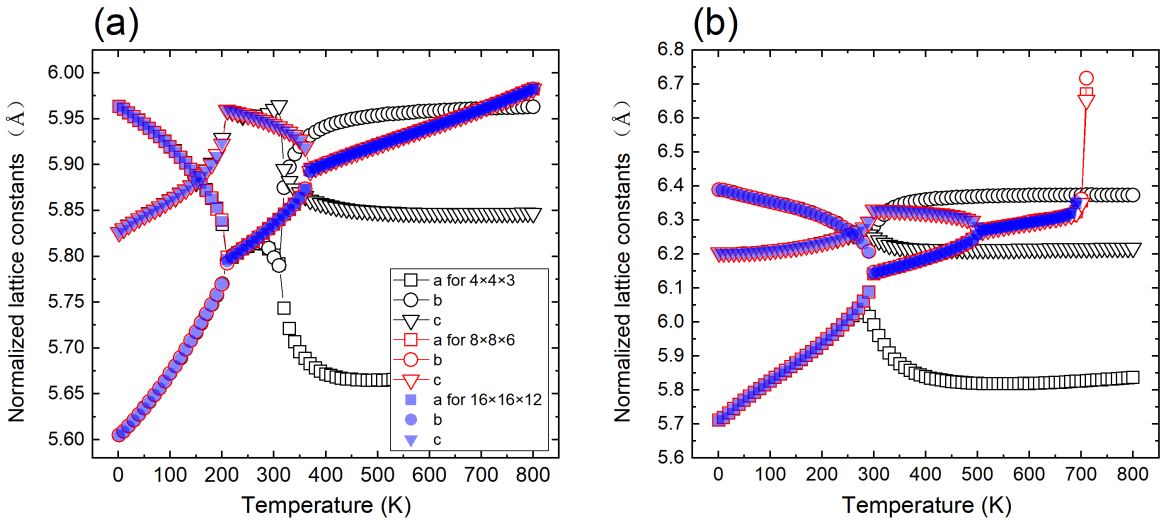

FIG. S1. Normalized lattice constants as a function of temperature. (a) $CsPbBr_3$, (b) $CsPbI_3$. NEP-based MD simulations are performed in $3 \times 3 \times 4$, $6 \times 6 \times 8$ and $12 \times 12 \times 16$ supercells.

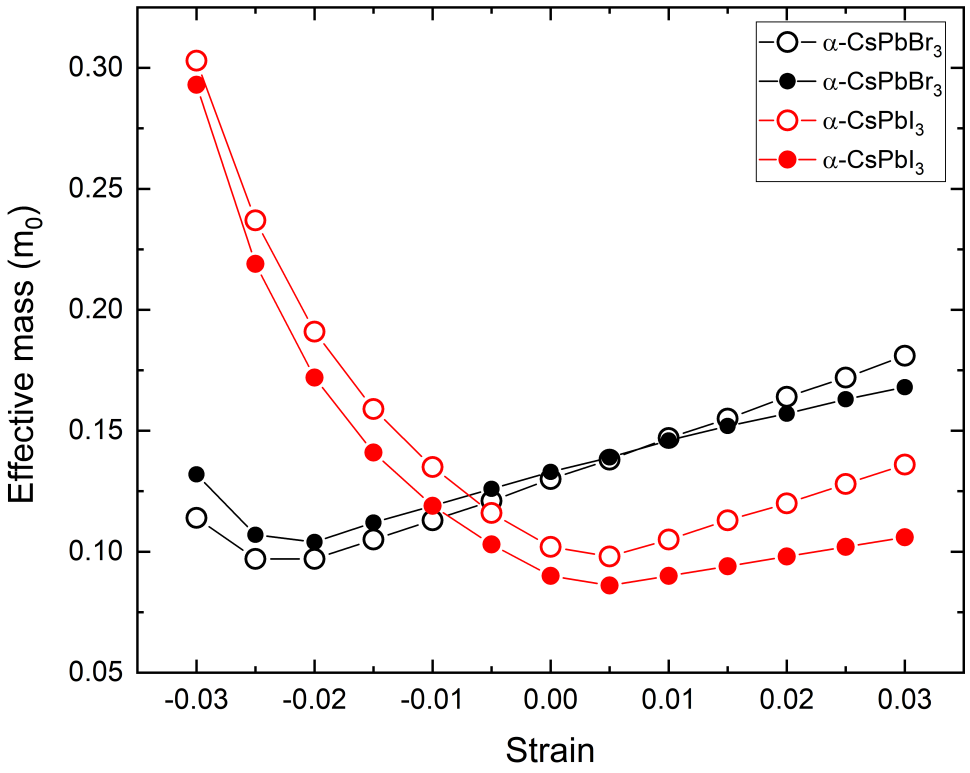

FIG. S2. Carrier effective mass as a function of lattice strain. Hollow and solid symbols represent hole and electron, respectively.

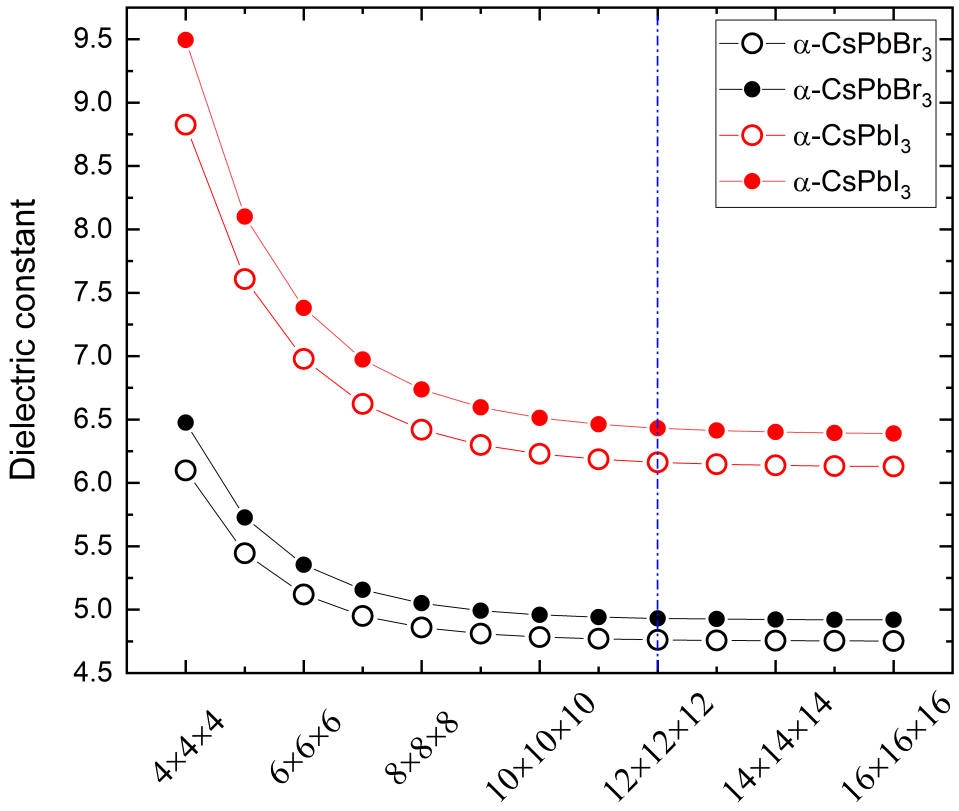

FIG. S3. Convergence test of dielectric constant for anharmonic perovskites. Hollow and solid symbols represent norm-conserving (NC) and projector augmented wave (PAW) pseudopotentials, respectively.

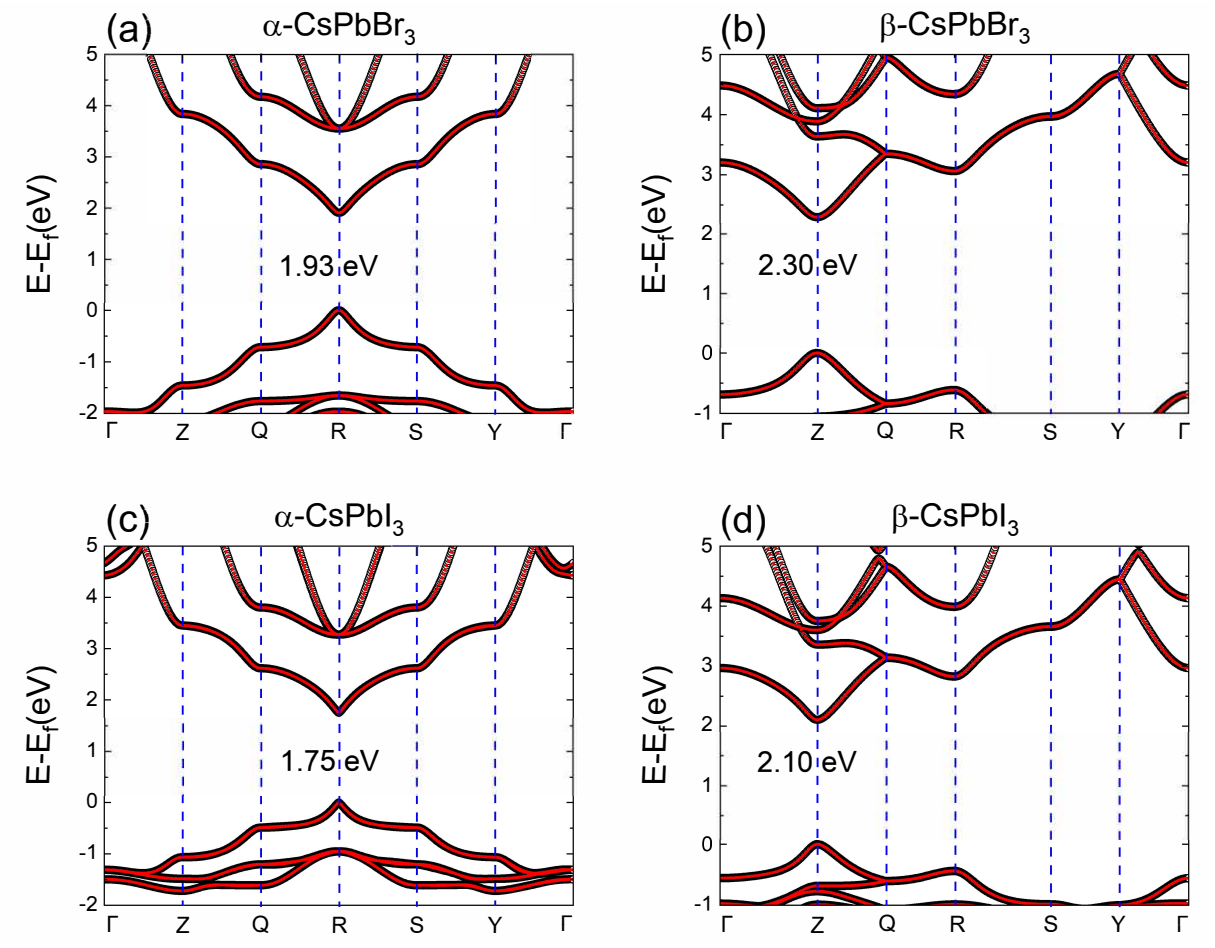

FIG. S4. Electronic band structure of anharmonic perovskite, which is calculated from both density functional theory (DFT) and Wannier function interpolation technique (WFIT). To correct the band edge shape, the spin orbit coupling (SOC) effect is taken into account in the calculation. The bandgap has been adjusted by HSE06 hybrid functional.

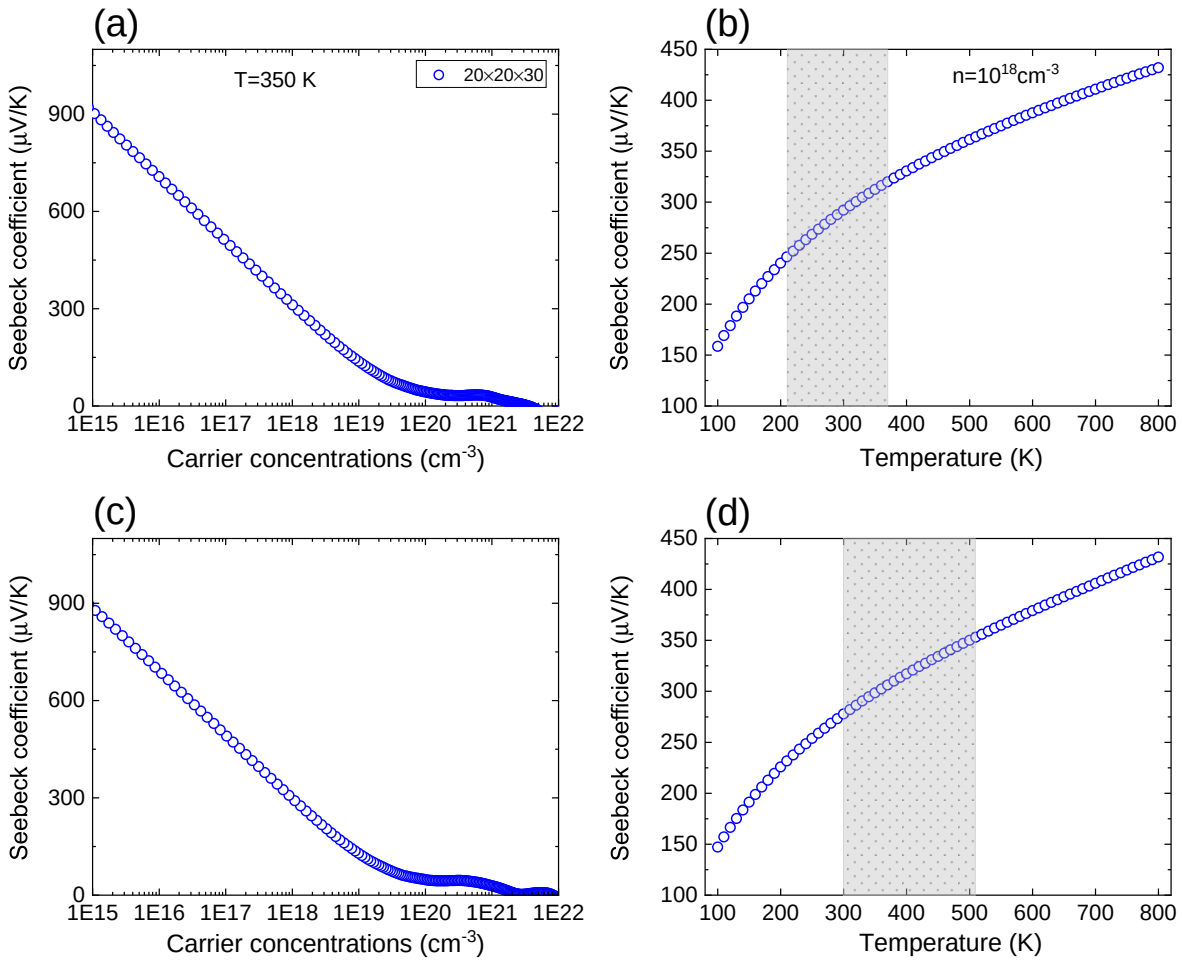

FIG. S5. Seebeck coefficient as a function of hole concentration and temperature. The gray region represents the tetragonal phase. (a) and (b): β-$CsPbBr_3$; (c) and (d): β-$CsPbI_3$.