Supporting Information for Impact of Composition Engineering on Charge Carrier Cooling in Hybrid Perovskites: Computational Insights

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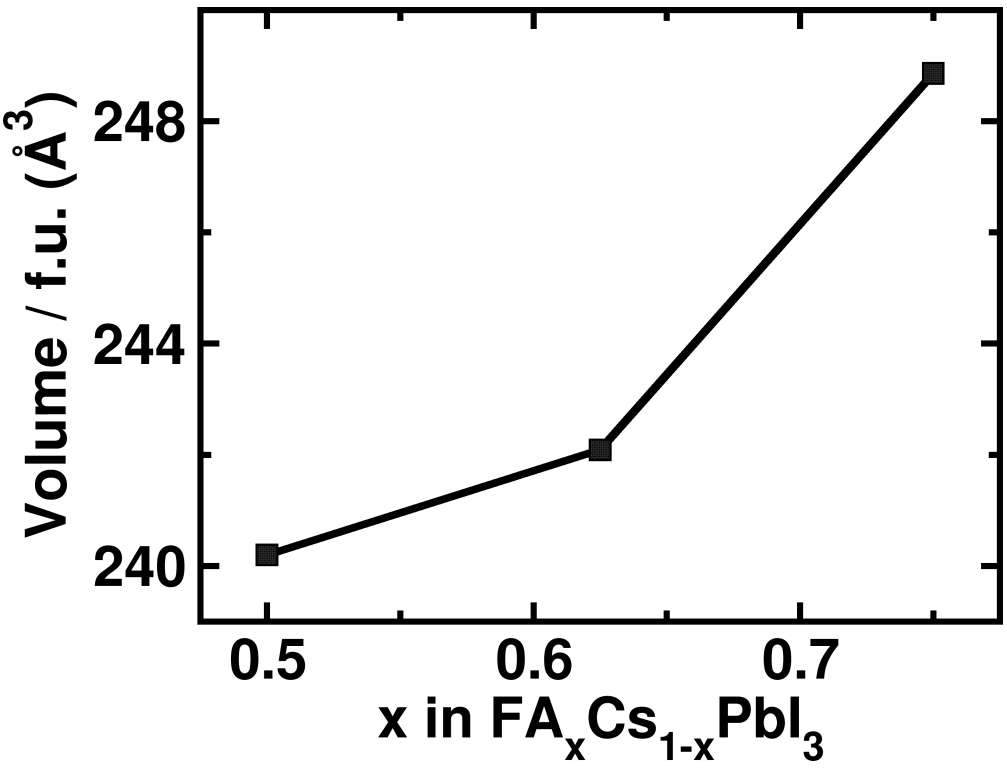

Figure S 1: The volume of $FA_{1-x}Cs_x$ with different Cs concentration. With increased Cs concentration the lattice volume decreases.

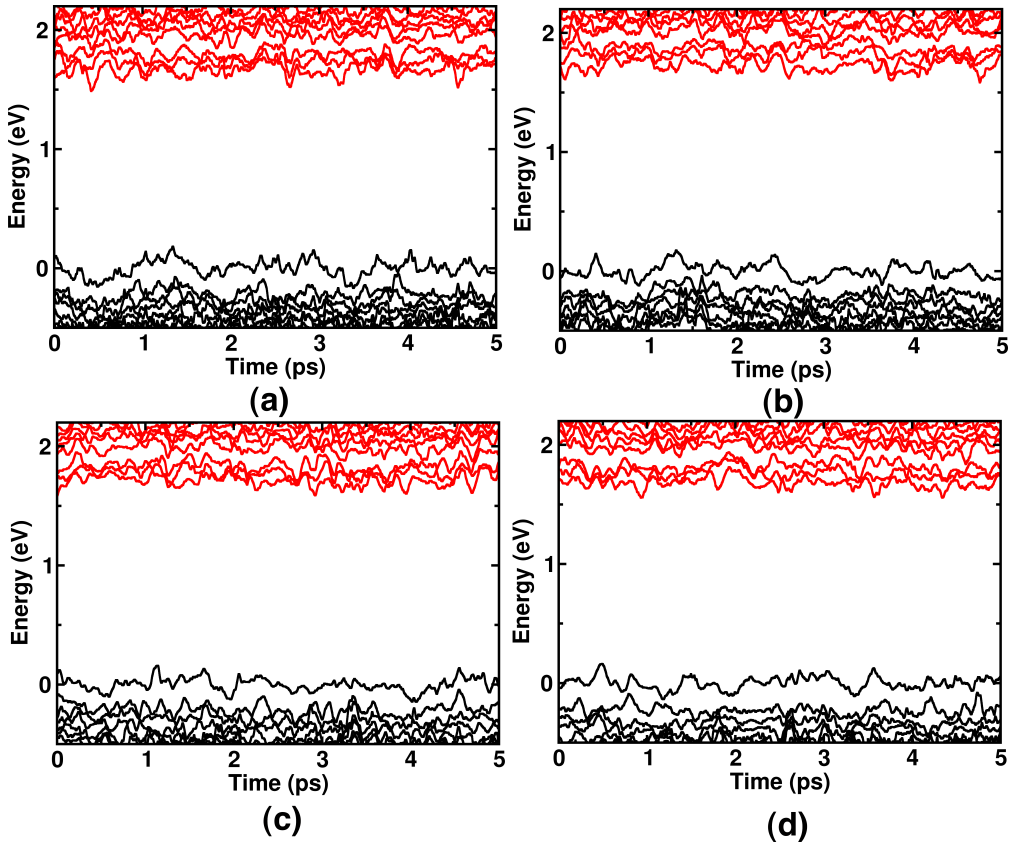

Figure S 2: The adiabatic time-dependent Kohn-Sham state energies during the AIMD simulation of 5 ps at 300 K for $FA_{1-x}Cs_x$ systems where (a) x = 0.0, (b) x = 0.25, (c) x = 0.375, and (d) x = 0.5. The energy states of these perovskites fluctuate due to the thermal vibrations in the lattice. We employ GGA-PBE functional-based DFT simulations to calculate these electronic structures.

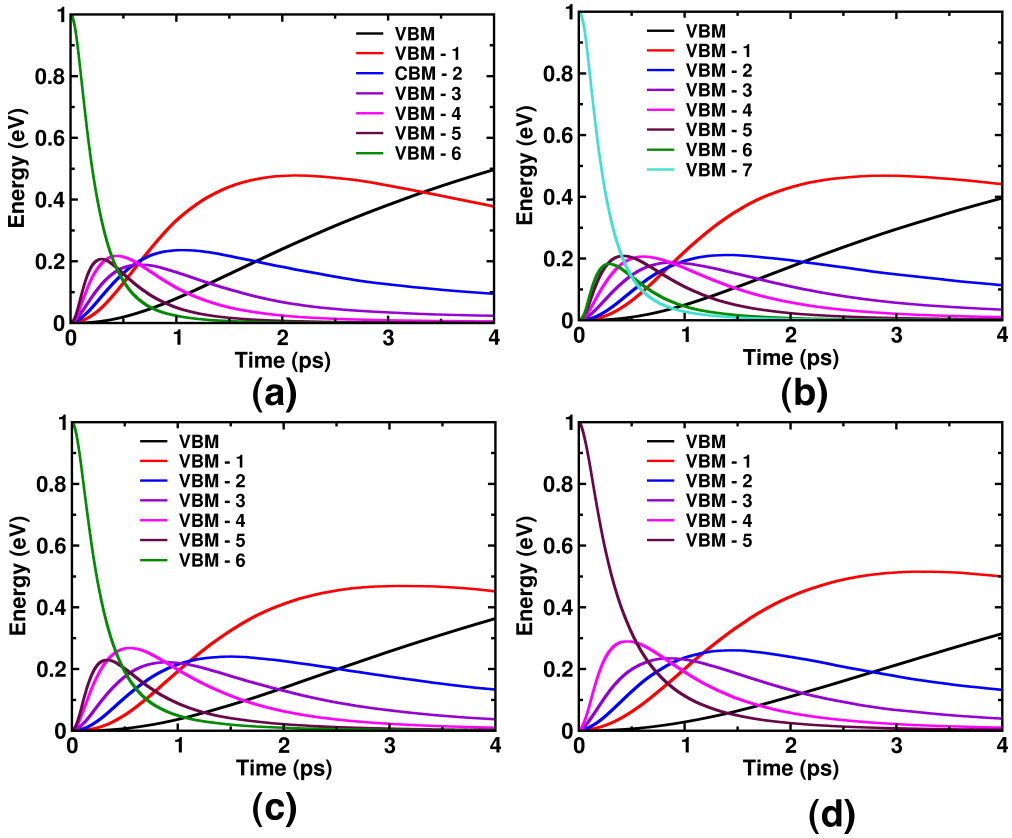

Figure S 3: The population of excited hole states over time in $FA_{1-x}Cs_x$ systems where (a) x = 0.0, (b) x = 0.25, (c) x = 0.375, (d) x = 0.5. With simulation time the cooled hole population increases at the band edge states (VBM, VBM-1).

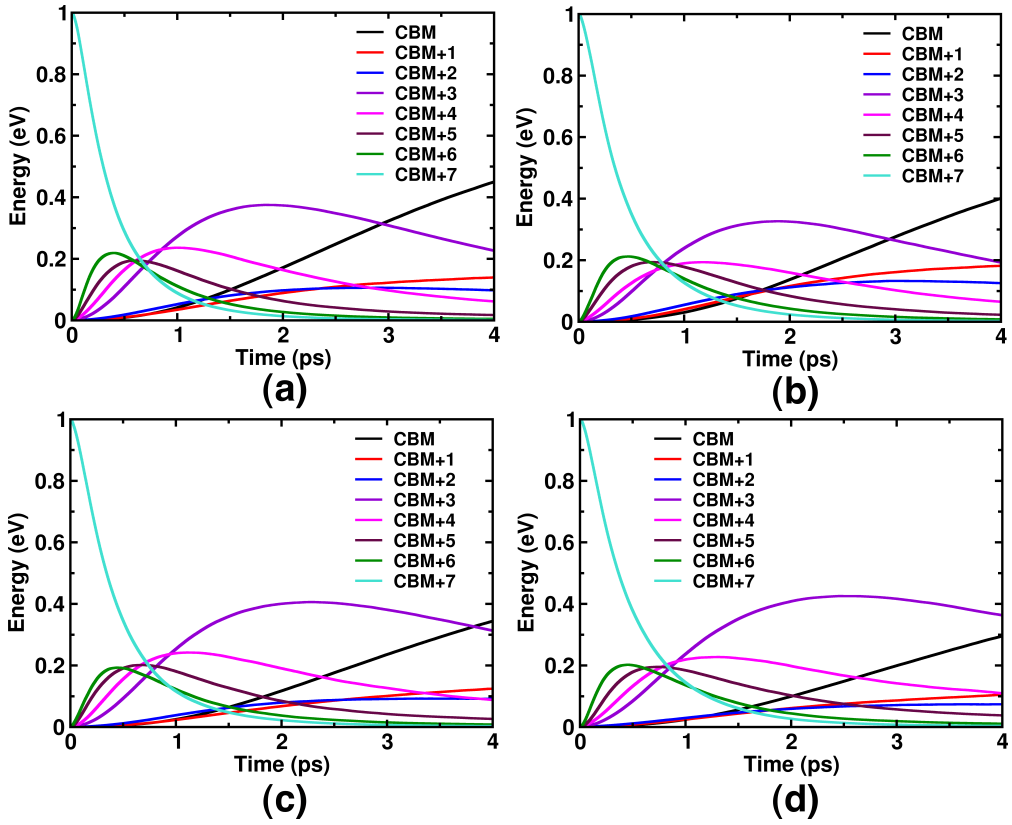

Figure S 4: The population of excited electron states over time in $FA_{1-x}Cs_x$ systems where (a) x = 0.0, (b) x = 0.25, (c) x = 0.375, (d) x = 0.5. The CBM+3 state contains significant amount of hot electron population initially. However, time the electron population further cools down to the band edge states (CBM, CBM+1).

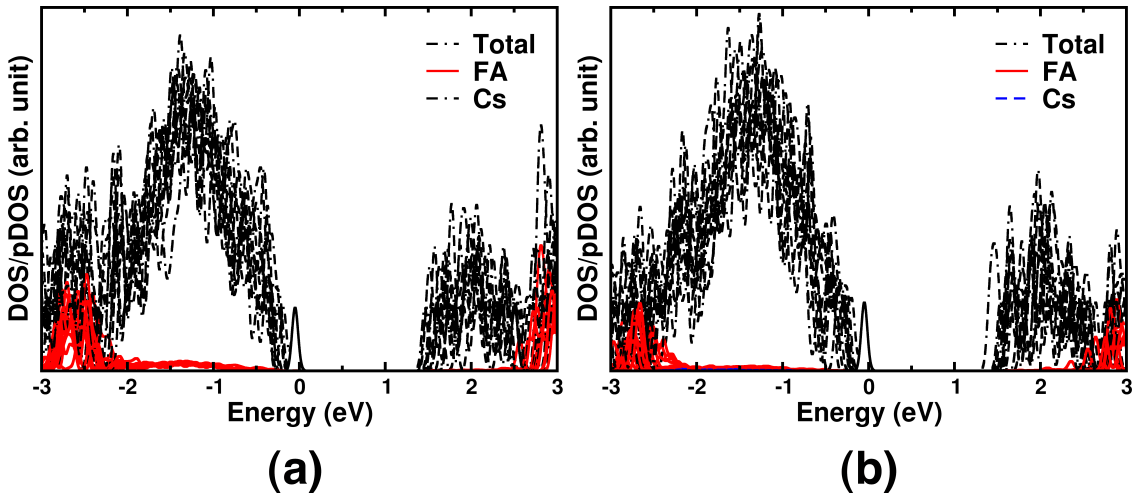

Figure S 5: The total and partial density of states of $FA_{1-x}Cs_x$ systems where (a) x = 0.0, (b) x = 0.5 over complete AIMD trajectories. We calculate electronic structures and density of states of these systems considering instantaneous geometries in every 50 fs for last 5ps of trajectories. The electronic states from A-cations remain away from band edge states over the dynamic trajectories.

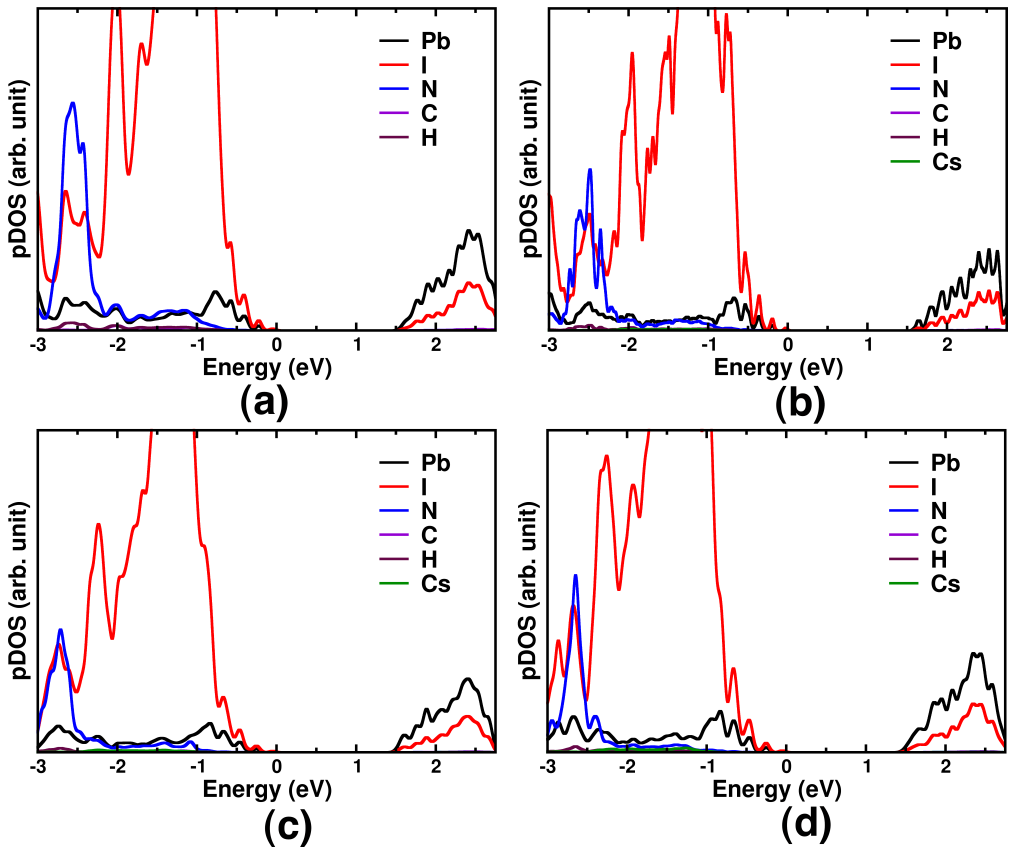

Figure S 6: The time-averaged magnitude of the carrier-phonon NACs in valence and conduction bands for $FA_{1-x}Cs_x$ systems where (a) x = 0.25, (b) x = 0.375. These NACs have been calculated using AIMD trajectories of last 5ps. In these plots, we represent i = 0 as the VBM, negative values are other deeper valence states, and positive values are indicating conduction band states.

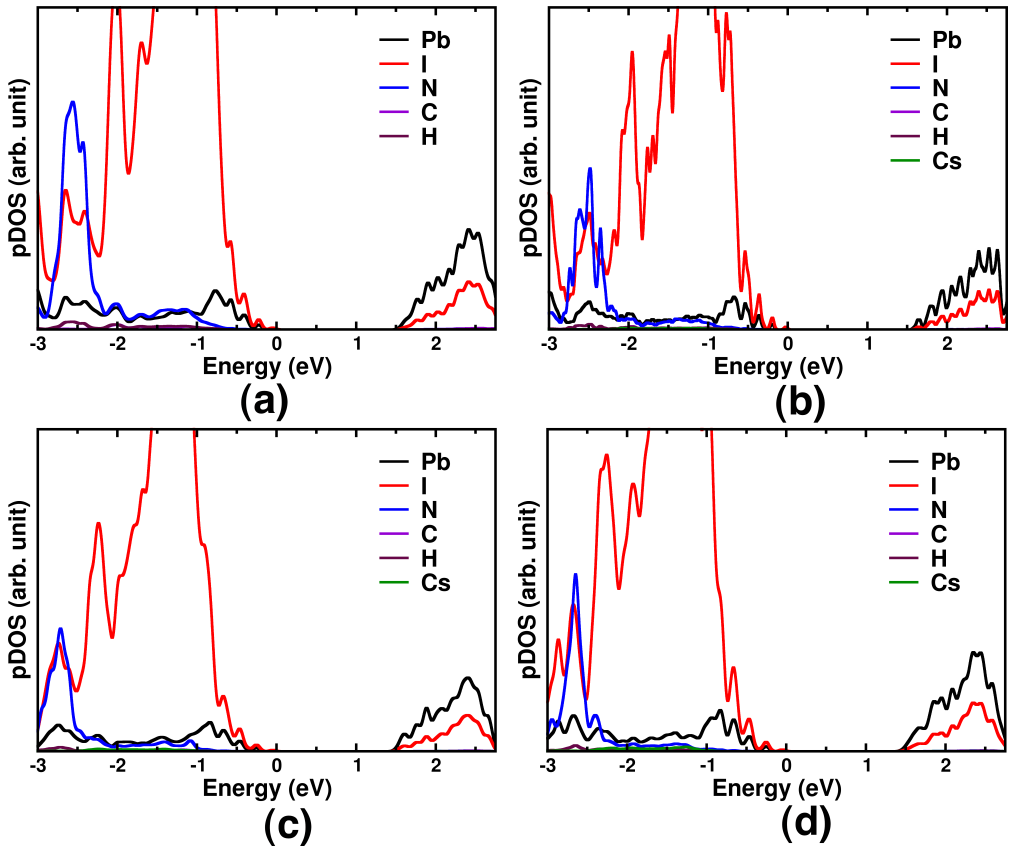

Figure S 7: The partial density of states (pDOS) in $FA_{1-x}Cs_x$ systems where (a) x = 0.0, (b) x = 0.25, (c) x = 0.375, (d) x = 0.5. These pDOS have been calculated using static optimized geometries and employing GGA-PBE functional.