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Local polar fluctuations in lead halide perovskites LIANG TAN, University of Pennsylvania, OMER YAFFE, Weizmann Institute of Science, YINSHENG GUO, LOUIS BRUS, Columbia University, ANDREW RAPPE, University of Pennsylvania, DAVID EGGER, LEEOR KRONIK, Weizmann Institute of Science — The lead halide perovskites have recently attracted much attention because of their large and growing photovoltaic power conversion efficiencies. However, questions remain regarding the temporal and spatial correlations of the structural fluctuations, their atomistic nature, and how they affect electronic and photovoltaic properties. To address these questions, we have performed a combined ab initio molecular dynamics (MD) and density functional theory (DFT) study on CsPbBr₃. We have observed prevalent anharmonic motion in our MD trajectories, with local polar fluctuations involving head-to-head motion of A-site Cs cations coupled with Br window opening. We calculate Raman spectra from the polarizability autocorrelation functions obtained from these trajectories and show that anharmonic A-site cation motion manifests as a broad central peak in the Raman spectrum, which increases in intensity with temperature. A comparison of the experimental Raman spectrum of hybrid organometallic MAPbBr₃ and fully inorganic CsPbBr₃ suggests that structural fluctuations in lead-halide perovskites is more general than rotation of polar organic cations and is intimately coupled to the inorganic framework.

Liang Tan
University of Pennsylvania

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