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Time Domain View of Liquid-like Screening and Large Polaron Formation in Lead Halide Perovskites[1] PRAKRITI PRADHAN JOSHI, KIYOSHI MIYATA, M. TUAN TRINH, XIAOYANG ZHU, Department of Chemistry, Columbia University — The structural softness and dynamic disorder of lead halide perovskites contributes to their remarkable optoelectronic properties through efficient charge screening and large polaron formation. Here we provide a direct time-domain view of the liquid-like structural dynamics and polaron formation in single crystal $CH_3NH_3PbBr_3$ and $CsPbBr_3$ using femtosecond optical Kerr effect spectroscopy in conjunction with transient reflectance spectroscopy. We investigate structural dynamics as function of pump energy, which enables us to examine the dynamics in the absence and presence of charge carriers. In the absence of charge carriers, structural dynamics are dominated by over-damped picosecond motions of the inorganic $PbBr_3^-$ sub-lattice and these motions are strongly coupled to band-gap electronic transitions. Carrier injection from across-gap optical excitation triggers additional ∼0.26 ps dynamics in $CH_3NH_3PbBr_3$ that can be attributed to the formation of large polarons. In comparison, large polaron formation is slower in $CsPbBr_3$ with a time constant of ∼0.6 ps. We discuss how such dynamic screening protects charge carriers in lead halide perovskites.

[1]US Department of Energy, Office of Science - Basic Energy Sciences

Prakriti Joshi
Columbia Univ

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