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Origins of Multivalley Electronic Transitions in Hybrid Perovskites Revealed by Transient Spectroscopy K. APPAVOO, Dept of Physics, UAB, W. NIE, J.C. BLANCON, Los Alamos National Laboratory, J. EVEN, Fonctions Optiques pour les Technologies de l'Information, CNRS, A.D. MOHITE, Los Alamos National Laboratory, M.Y. SFEIR, Center for Functional Nanomaterials, BNL — Mapping complex electronic excitations to the intricate lattice structure of hybrid organic-inorganic perovskites is critical to understand charge separation and hot-carrier extraction, processes that dictate energy-conversion and optoelectronic technologies. Here, we highlight how the dipolar CH3NH3+ organic molecule interacts with the inorganic PbI6- octahedral cage to impact the multiband, multivalley electronic structure of this halide perovskite. This is achieved by tracking the transient broadband optical spectra while tuning the structural lattice of the hybrid perovskite via its reversible temperature-dependent phase transition (PT). These temperature-dependent optical snapshots, here captured at 5 ps, reveal exquisite details of those bands, reporting for the first time a degeneracy lifting in the tetragonal state at 2.6 eV that increases as the organic molecule rotational degrees of freedom are suppressed in the orthorhombic state. Plotting this dispersion relation, along with a symmetry analysis of the PT, we describe how the electronic states evolve from the tetragonal to orthorhombic phase and ascribe the splitting to the nearly degenerate transitions at the R and M points of the Brillouin zone. Furthermore, a zone folding in the orthorhombic state explains other salient features of our experiments, such as the emergence of other allowed transitions near 2 eV and a decrease in hot carrier lifetime.

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