Hybrid inorganic-organic halide perovskite crystals, such as CH$_3$NH$_3$PbI$_3$, have garnered significant attention for their use as the light-absorbing semiconductor layer in low-cost photovoltaic devices with high power conversion efficiencies (>20%). The performance of these materials challenges conventional wisdom in that these materials exhibit excellent electrical performance despite the inexpensive, low-temperature, solution-based preparatory routes that result in imperfect materials with small grain sizes. Therefore, we have focused on (a) understanding the fundamental origins of defect tolerance in these semiconductors and (b) establishing a relationship between the dynamics of organic molecules in the crystal structures and the functional electronic properties. Our results - which include in-depth synchrotron X-ray scattering and neutron spectroscopy - provide a platform from which to understand structure-dynamics-property relationships in functional perovskite halides, which have the potential for transformative applications in photovoltaics.