Light-induced picosecond rotational disordering of the inorganic sublattice in hybrid perovskites

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Femtosecond resolution electron scattering techniques are applied to resolve the first atomic-scale steps following absorption of a photon in the prototypical hybrid perovskite methylammonium lead iodide. Following above-gap photo-excitation, we directly resolve the transfer of energy from hot carriers to the lattice by recording changes in the root-mean-square atomic displacements, on ten picosecond time-scales. Measurements of the time-dependent pair distribution function show an unexpected broadening of the iodine-iodine correlation function while preserving the Pb-I distance. This indicates the formation of a rotationally-disordered halide octahedral structure developing on picosecond time-scales. This work shows the important role of light-induced structural deformations within the inorganic sublattice in elucidating the unique optoelectronic functionality exhibited by hybrid perovskites and provides new understanding of hot carrier – lattice interactions which fundamentally determine solar cell efficiencies.