

# Photo-driven dipole reordering: key to carrier separation in metalorganic halide perovskites

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Photo-driven dipole reordering of the intercalated organic molecules in halide perovskites has been suggested to be a critical degree of freedom, potentially affecting physical properties, device performance, and stability of hybrid perovskite-based optoelectronic devices. However, thus far a direct atomically-resolved dipole mapping under device operation condition, i.e. illumination, is lacking. Here, we map simultaneously the molecule dipole orientation pattern and the electrostatic potential with atomic resolution using light-illuminated cross-sectional scanning tunneling microscopy and spectroscopy.

Our experimental observations demonstrate that a photo-driven dipole reordering, initiated by a photo-excited separation of electron-hole pairs in spatially displaced orbitals, leads to a fundamental reshaping of the potential landscape in halide perovskites, creating separate one-dimensional transport channels for holes and electrons. We anticipate that analogous light-induced polarization order transitions occur in bulk and are at the origin of the extraordinary efficiencies of organometal halide perovskite-based solar cells as well as could reconcile apparently contradictory materials' properties.

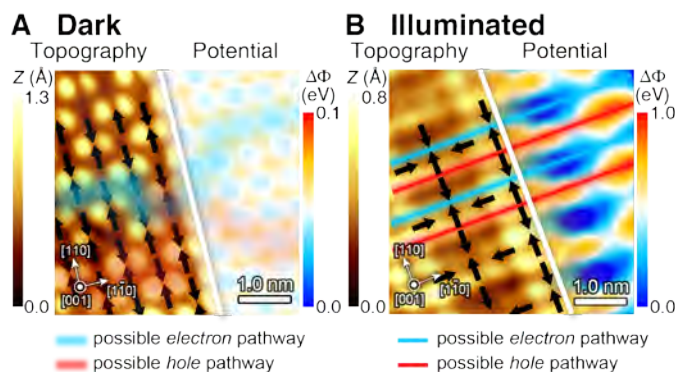


Figure 1 Illustration of light-induced structure and potential changes at  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  surfaces. (A)  $2\times 2$  phase in dark and (B) illumination-induced  $4\times 2$  phase. Superimposed are the constant-current STM images (left side) and electrostatic potential energy landscapes (right).

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