

Supplementary Materials

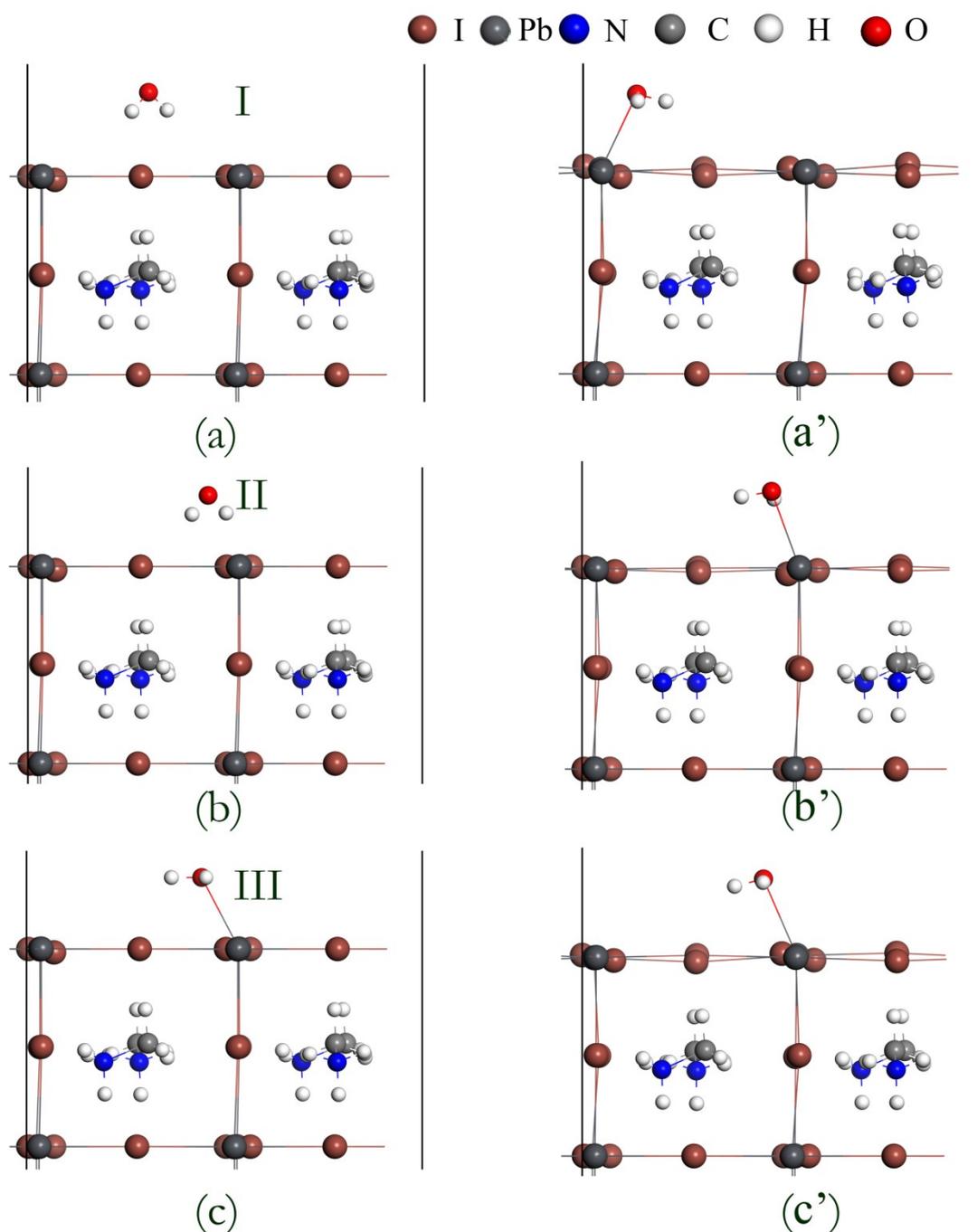


Fig.S1. Structures with adsorbed H₂O molecules on surfaces of photo-dilated MAPbI₃ lattice with Pb-I-Pb angle of 166.08° (β phase). (a) Position *I*, i.e., H₂O just lies above the hollow site surrounded by the Pb-I square. (b) Position *II*, where H₂O molecule locates near a corner of Pb-I square with two hydrogen atoms in H₂O molecule pointing downward. (c) Position *III*, i.e., H₂O is also near a corner of Pb-I square, but with H₂O molecule paralleling to the PbI surface. (a'), (b') and (c') are configurations after relaxations, respectively.

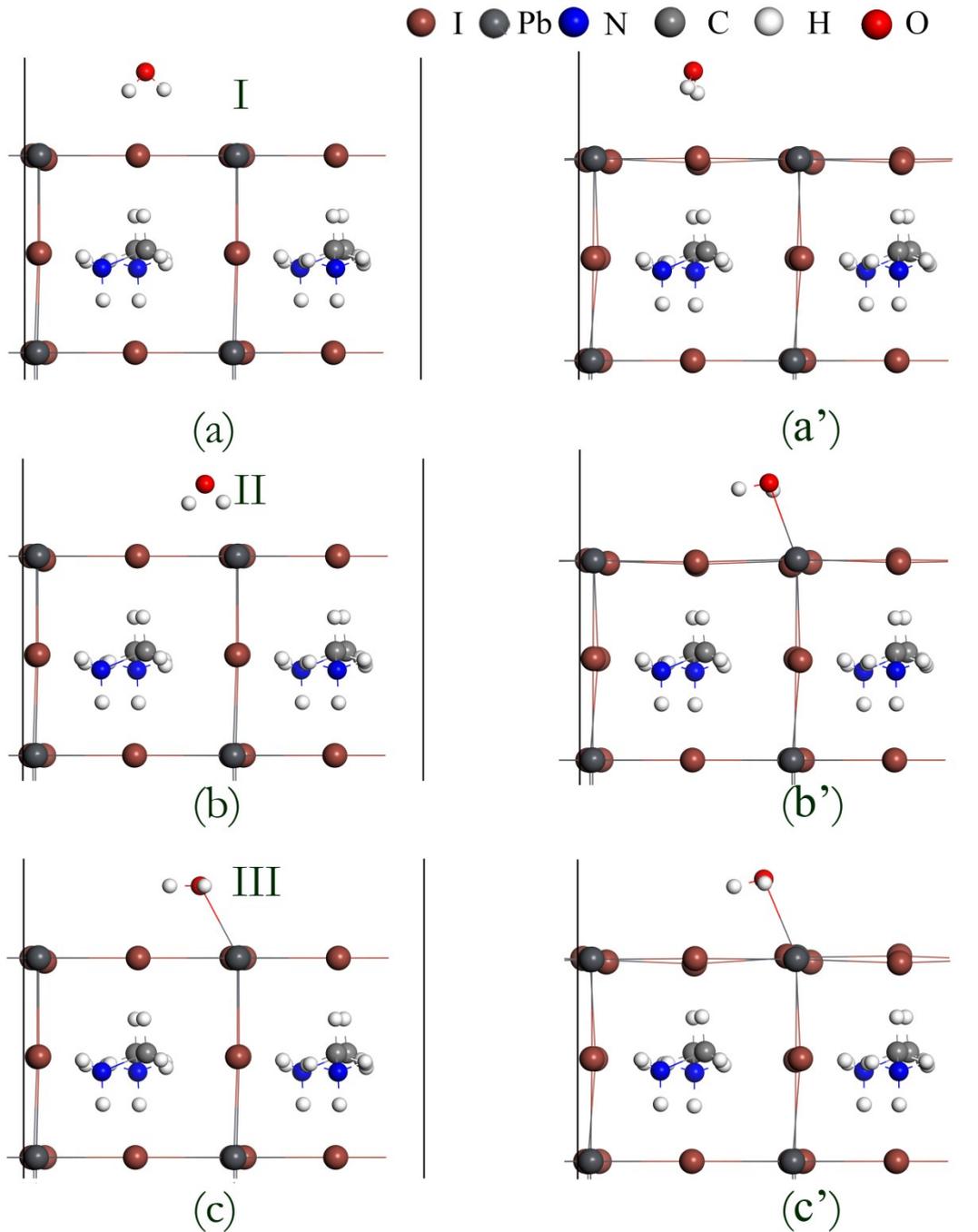


Fig.S2. Structures with adsorbed H₂O molecules on surfaces of photo-dilated MAPbI₃ lattice with Pb-I-Pb angle of 173.08° (γ phase). (a) Position **I**, i.e., H₂O just lies above the hollow site surrounded by the Pb-I square. (b) Position **II**, where H₂O molecule locates near a corner of Pb-I square with two hydrogen atoms in H₂O molecule pointing downward. (c) Position **III**, i.e., H₂O is also near a corner of Pb-I square, but with H₂O molecule paralleling to the PbI surface. (a'), (b') and (c') are configurations after relaxations, respectively.

