

## **Description of Additional Supplementary Files**

### **File Name: Supplementary Movie 1**

**Description:** The interfacial preference of  $\text{PbI}_2$  nanosheet, the video was generated by classical molecular dynamics simulation. The monolayer  $\text{PbI}_2$  nanosheet will spontaneously move toward the interface, where the  $\text{PbI}_2$  nanosheet is initially placed in bulk water. The snapshot structures can be obtained from Fig. 2e.

### **File Name: Supplementary Movie 2**

**Description:** Molecular dynamics simulations of  $\text{PbI}_2$  growth at the water-air interface. The video was generated through classical molecular dynamics simulation. The small grey and yellow spheres represent the Pb and I atoms of a pre-existing  $\text{PbI}_2$  nanosheet, respectively. The large grey and yellow spheres represent the Pb and I ions initially dissolved in water, respectively. Snapshot structures can be referenced in Fig. 2f.

### **File Name: Supplementary Movie 3**

**Description:**  $\text{PbI}_2$  growth process in its native environment at 298 K.