# Controlled formation of ferroelectric proton ordering in ice films

Jiani Hong[1,2#], Tiancheng Liang[1], Yuejian Zhang[1,2], Ye Tian[1,2*], Li-Mei Xu[1,2*], En-Ge Wang[1,2*] and Ying Jiang[1,2*]

[1] *International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, P. R. China*

[2] *Interdisciplinary Institute of Light-Element Quantum Materials and Research Center for Light-Element Advanced Materials, Peking University, Beijing 100871, P. R. China*

# timeless@pku.edu.cn

Water possesses intrinsically large molecular electric dipole moments, and proton ordering within hydrogen-bond network is fundamental to diverse phenomena in materials science, electronics, biology and astrophysics. However, achieving controlled proton ordering in condensed phases remains challenging, with the underlying microscopic mechanisms still elusive. Recently, we developed a hydrogen-sensitive scanning probe microscopy (SPM) technique based on higher-order electrostatic forces [1], which enabled successful applications to interfacial water and ice systems [2,3]. Subsequently, we further established a universal high-resolution imaging method for insulating surfaces, allowing direct visualization of bulk hexagonal ice (ice Ih) surfaces [4]. These advances open up new possibilities for probing proton ordering in ice with atomic-scale spatial resolution. Here, we apply this approach to resolve the orientations of individual water molecules during the layer-by-layer growth of ice films and determine interfacial polarization. Combined with calculations, our results reveal that minimal proton doping induces a two-dimensional (2D) proton-ordered ice XI phase across diverse metal substrates, which subsequently serves as a template for the growth of bulk ice XI. These findings uncover the atomic-scale mechanism underlying the formation and stabilization of ferroelectric proton ordering at interfaces.

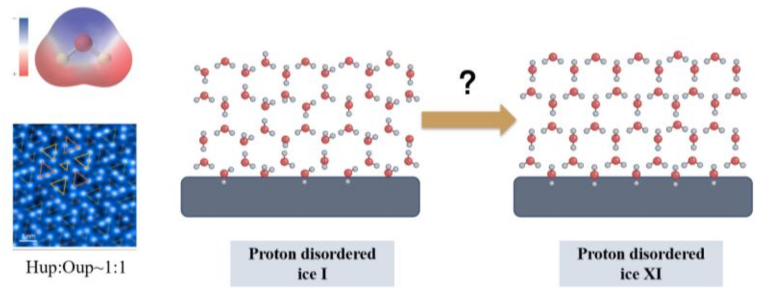


**Figure 1.** Schematic illustration of water molecule polarization, proton-ordered ice Ih, and proton-disordered ice XI.

Reference

[1] Jinbo Peng, et al. Nat. Commun. **9**, 122 (2018).

[2] Runze Ma, et al. Nature **577**, 60 (2020).

[3] Ye Tian, et al. Science **377**, 315 (2022).

[4] Jiani Hong, et al. Nature **630**, 375 (2024).