In-situ TEM study of ice nucleation and growth

Lifen Wang[1]
[1]Institute of Physics, Chinese Academy of Sciences, Zhongguancun, China

Ice is one of the most omnipresent solids on Earth, yet its crystallization is not fully understood. In particular, a long-standing debate is whether natural ice can form in a metastable pure-cubic phase. A mainstream view attributes the divergence to the stacking disordered ice (Isd) with a mixture of cubic and hexagonal sequences that preferentially nucleate and grow at low temperatures typically under 200 K, where complex kinetics in ice crystallization has also defied understanding thus far. This is due to that the direct and reliable determination of the structure of metastable ice requires ultra-high spatiotemporal resolution in an in-situ freezing environment, which remains a major outstanding challenge in the community. Here, through developing the in-situ cryogenic TEM, we directly tracked and unveiled the heterogeneous polymorphic ice nucleation and crystallization with molecular resolution during the water vapor deposition process at 102 K. We further pinpointed the atomic structure of polymorphic ice crystallites and identified distorted ice tetrahedral configurations as crystal defects with molecular resolution, which were not accessible before. Employing the electron beam as both the imaging probe and activation source, we were able to trigger and consequently monitor the kinetic transition process of stacking disordered ice (Isd) to ice Ic, demonstrating the unexpected meta-stability of ice Isd with respect to ice Ic. Our measurements shed new light on the structure and dynamics process determined heterogeneous nucleation of polymorphic ice. While a deep understanding of ice crystallization requires microscopic details, our realization of direct real-space imaging of growing ice crystallites at the molecular level is a key step towards reconciling experimental and theoretical investigations into the structure, nucleation, and crystallization based complex phenomena in ice family and polymorphic crystals.

Keywords:

Ice, In-situ TEM, polymorphism, Crystallization

Reference:

1 Huang, X. et al. Tracking cubic ice at molecular resolution. Nature 617, 86-91, doi:10.1038/s41586-023-05864-5 (2023).
2 Wang, L. et al. Microscopic Kinetics Pathway of Salt Crystallization in Graphene Nanocapillaries. Physical Review Letters 126, 136001, doi:10.1103/PhysRevLett.126.136001 (2021).
3 Wang, L. et al. Synthesis of Honeycomb-Structured Beryllium Oxide via Graphene Liquid Cells. Angewandte Chemie International Edition 59, 15734-15740, doi:https://doi.org/10.1002/anie.202007244 (2020).
4 Zhu, L. et al. Visualizing Anisotropic Oxygen Diffusion in Ceria under Activated Conditions. Physical Review Letters 124, 056002, doi:10.1103/PhysRevLett.124.056002 (2020).