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Observing growth and interfacial dynamics of nanocrystalline ice in thin amorphous ice films

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Ice crystals at low temperatures exhibit structural polymorphs including hexagonal ice, cubic ice, or a hetero-crystalline mixture of the two phases. Despite the significant implications of structure-dependent roles of ice, mechanisms behind the growths of each polymorph have been difficult to access quantitatively. Using in-situ cryo-electron microscopy and computational ice-dynamics simulations, we directly observe crystalline ice growth in an amorphous ice film of nanoscale thickness, which exhibits three-dimensional ice nucleation and subsequent two-dimensional ice growth. We reveal that nanoscale ice crystals exhibit polymorph-dependent growth kinetics, while hetero-crystalline ice exhibits anisotropic growth, with accelerated growth occurring at the prismatic planes. Fast-growing facets are associated with low-density interfaces that possess higher surface energy, driving tetrahedral ordering of interfacial H_2O molecules and accelerating ice growth. These findings, based on nanoscale observations, improve our understanding on early stages of ice formation and mechanistic roles of the ice interface.

Ice crystallization is a ubiquitous process having significant implications in various scientific and technological fields[1–7]. The formation of structural polymorphs, including amorphous and crystalline forms of ice are influenced by conditions such as temperature, pressure, and preparation protocols[4,8–19]. Hexagonal ice (ice I_h), the most common crystalline polymorph, is widely found in atmospheric conditions[11,15]. Cubic ice (ice I_c) is another crystalline polymorph reported to coexist with ice I_h at a broad temperature range of 160–240 K[13,15]. Despite the marginal difference in the thermodynamic favorability for the growths of ice I_c and I_h[20,21], these polymorphs exhibit distinct kinetic growth properties[22,23]. This implies that there are other important factors that influence the nucleation and growth of ice crystals. Considering the important role of interfaces in the nucleation and growth of solids[24–29], factors affecting ice growth are also likely relevant to interfacial characteristics of ice polymorphs[12,13]. Indeed, there had been observations in which ice growth shows a dependency on the nature of interfaces in the environment. For instance, conditions within nanopores[30] or nanoscale water droplets[22,31] have favored the formation of ice I_c structures over I_h.

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