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## Acid-Promoted Crystallization of Amorphous Solid Water

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Amorphous solid water (ASW) has attracted much attention in the ice research community because of its unique properties as a metastable solid form of water and possible existence in interstellar clouds. ASW is kinetically metastable and undergoes a phase transition to crystalline ice (CI) upon heating; these phenomena have been studied in depth in recent years. [1-3] In this presentation, we discuss the effect of excess protons (hydroniums) on the crystallization of ASW. We prepared an ASW film sample by water vapor deposition on a Pt(111) single crystal surface in an ultra-high vacuum environment. Excess protons were generated by the ionization of adsorbed hydrogen chloride (HCl) gas on the ASW surface. The crystallization of an ASW film was monitored by temperature-programmed desorption and reflection-absorption infrared spectroscopy measurements. In contrast to a normal behavior that solute species retard the crystallization of ASW due to an entropic effect, the excess protons released from HCl molecules facilitated the crystallization of ASW. This phenomenon was observed when HCl was supplied not only at the surface of an ASW film but also in the interior region. In both cases, the crystallization was initiated at the location where the acid was provided to the sample. The presence of excess protons lowered the apparent activation energy of the crystallization of ASW. A possible promotion mechanism for the crystallization of ASW is discussed in relation to the properties of excess protons in ASW.

[1] Safarik, D. J.; Mullins, C. B. The nucleation rate of crystalline ice in amorphous solid water. *J. Chem. Phys.* (2004) 121, 6003-6010.

[2] Backus, E. H.; Grecea, M. L.; Kleyn, A. W.; Bonn, M. Surface crystallization of amorphous solid water. *Phys. Rev. Lett.* (2004) 92, 236101.

[3] Yuan, C.; Smith, R. S.; Kay B. D. Surface and bulk crystallization of amorphous solid water films: Confirmation of “top-down” crystallization. *Surf. Sci.* (2016) 652, 350-354.

### Significance statement

A solute species usually suppresses the crystallization of aqueous solution due to an entropic effect. However, a contrasting effect was observed for hydrogen chloride in ASW, which promotes the crystallization of ASW. This unique phenomenon brings attention to the ASW crystallization mechanism in relation to the behavior of excess protons.

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