Deciphering the morphology of ice films on metal surfaces

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Although extensive research has been aimed at the structure of ice films [1,2], questions regarding basic processes that govern film evolution remain. Recently we discovered how ice films as many as 30 molecular layers thick can be imaged with STM [3]; for thicker films AFM has to be used. The observed morphology yields new insights about water-solid interactions and how they affect the structure of ice films. This talk gives an overview of this progress for crystalline ice films on Pt(111) [3-8]. STM reveals a first molecular water layer very different from bulk ice: besides the usual hexagons it also contains pentagons and heptagons [4]. Slightly thicker films (~1nm, at T>120K) are comprised of ~3nm-high crystallites, surrounded by the one-molecule-thick wetting layer (Figure 1). These crystals dewet by nucleating layers on their top facets [5]. Measurements of the nucleation rate as a function of crystal height provide estimates of the energy of the ice-Pt interface. For T>115K surface diffusion is fast enough that surface smoothing and 2D-island ripening is observable [6]. By quantifying the T-dependent ripening of island arrays we determined the activation energy for surface self-diffusion. The shape of these 2D islands varies strongly with film thickness. We attribute this to a transition from polarized ice at the substrate towards proton disorder at larger film thicknesses [7]. Despite fast surface diffusion ice multilayers are often far from equilibrium. For example, the crystal structure of ice, deposited at low temperatures (~140 K), switches twice as films grow thicker. Isolated 3D clusters, which can only grow via layer nucleation, consist of hexagonal ice. Following coalescence, cubic ice is being produced in growth spirals created by screw dislocations above substrate steps [3]. Eventually, at thicknesses of ~20 nm, a different type of growth spiral, generated by dislocations with a double burgers vector, becomes dominant causing the preferential formation of hexagonal ice [8].

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Figure 1:

2-3 nm thick ice crystals grown at 140 K onto Pt(111) and imaged non-destructively with STM.