Deciphering the morphology of ice films on metal surfaces

Konrad Thürmer, Sandia National Laboratories, Livermore, CA, U.S.A.

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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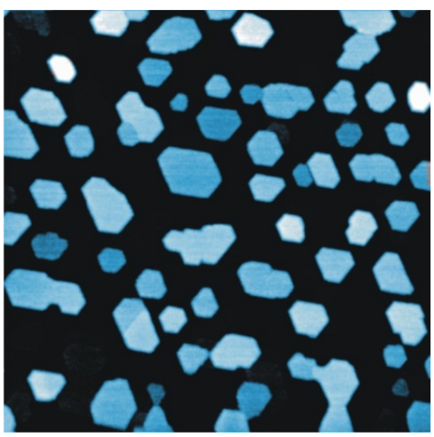

Figure 1:
2-3 nm thick ice crystals grown at 140 K onto Pt(111) and imaged non-destructively with STM.

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