A roughening transition on the surface of ice.

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The habit of ice crystals in the atmosphere change from plates, to columns, to plates and yet back to columns as temperature is cooled down below the triple point [1]. Attempts to explain this puzzling sequence of events rely on the formation of a thin quasi-liquid layer of premelted ice [2]. Many efforts have been devoted to determine the onset of premelting and the thickness of the layer as the triple point is approached [3]. But precisely what is the influece of this film on the global behavior of the ice/vapor interface, and how could it impact on the mechanism of crystal growth is far from being understood [1]. In this paper, we argue that a thin premelting layer of ice hardly one nanometer thick is able to induce a structural transition of the Kosterlitz-Thoules type on the ice surface.[4] Our computer simulations reveal that the two distinct surfaces bounding the quasi-liquid layer behave at small wave-lengths as rough and independent ice/water and water/vapor interfaces. However, the finite thickness of the layer inhibits large scale fluctuations and drives the crystal surface smooth at long wave-lengths. Our results explain why ice crystal prisms retain a distinct hexagonal shape up to the triple point, and suggest the formation of a premelting film could slow down the growth rate of crystal facets. Understanding the structure and growth mechanisms of ice crystals also has important implications in atmospheric science, glaciology, and frost heaving [3].

In our study, we simulate the premelting layer of water a few Kelvin below the triple point. Using an adequate order parameter, it is possible to identify distinct ice/film and film/vapor, surfaces, which separate the premelting film from the bulk solid and vapor. The spectrum of surface fluctuations allow us to meassure the wave-vector dependent components of the stiffness tensor, which are finite for a rough surface, but effectively diverge for smooth surfaces.

At a temperature two Kelvin below the triple point, our results (Fig.1) indicate that the stiffness coefficients for ice/film (blue), film/vapor (red) and coupled ice/film and film/vapor (green) fluctuations converge to a finite value. Moreover, it is found that the fluctuations closely resemble those of independent ice/water and water/vapor interfaces (empty symbols) at large wave-vectors, but eventually couple and produce an effective stiffness which is the sum of the stiffness coefficients of the independent interfaces (black arrow). A few Kelvin below, however, the stiffness coefficients effectively diverge, and indicate the onset of a completely different regime with finite surface fluctuations.

The roughening transition that is observed may be explained using a Capillary Wave Hamiltonian to describe the liquid/vapor fluctuations and a sine Gordon model to mimick the solid/liquid interface. Both Hamiltonians are coupled via an interface potential. The solution of the model (full lines) indicates that the presence of a premelting film has the effect of smoothening the ice surface well above the roughening transition of the ice/water interface. As a result, it is expected that the crystal growth rate of the prismatic facet will slow down below this temperature, and promote the growth of columnar crystals as observed in the atmosphere.

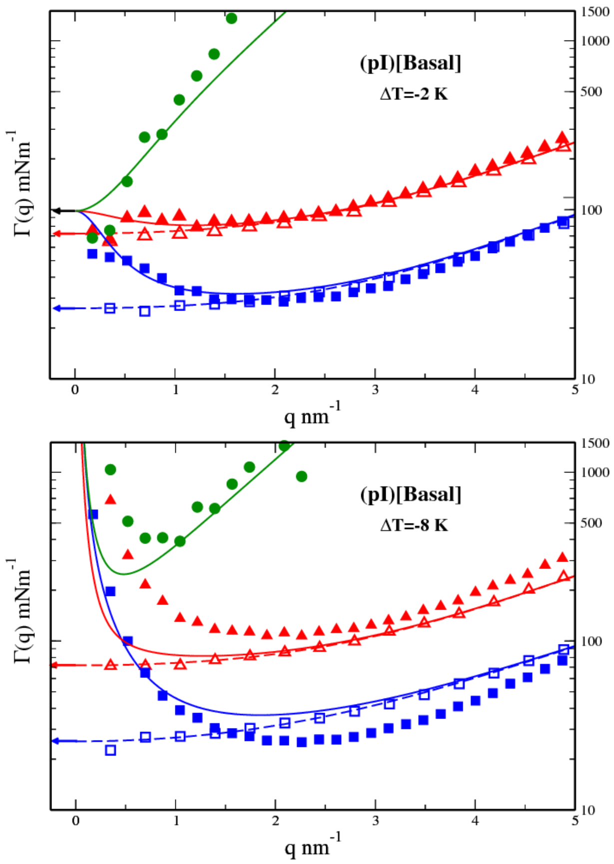

Figure 1: Observation of a roughening transition on the prismatic surface of ice. At a temperature 2 K below the triple point, the wave-vector dependent components of the stiffness tensor converge to a finite value at $q \to 0$ that is the sum of the ice/water and water/vapor stiffness coefficients. Six Kelvin below, the stiffness coefficients diverge, indicating the onset of a smooth facet.

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