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 \cite{1}. Attempts to explain this puzzling sequence of events rely on the formation of a thin quasi-liquid layer of premelted ice \cite{2}. Many efforts have been devoted to determine the onset of premelting and the thickness of the layer as the triple point is approached \cite{3}. But precisely what is the influence 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 \cite{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.\cite{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.

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 measure 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, indicating the onset of a smooth facet 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.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{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 $q \rightarrow 0$ 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.}
\end{figure}

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