Surface phase transitions, anomalous step free energies and crystal growth rates of ice in the atmosphere

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As the thread of global warming is confirmed, the fate of ice has become a serious concern. Unfortunately, our understanding of how ice actually grows (or melts) remains poor. The Nakaya diagram, which describes the growth forms of snow crystals in the atmosphere provides an eloquent example [1]. At low vapor saturation, ice crystallites are found as simple hexagonal prisms, but their shape changes from plates, to columns to plates and yet back to columns as temperature is cooled down below 0 C. Experiments allow to correlate empirically these shape transitions with a crossover of crystal growth rates, but the physics behind these transformations and its relation with the underlying equilibrium surface structure remains completely unknown.

In this work we exploit a methodology for the study of surface fluctuations developed in the last years in our lab [2, 3, 4, 5] to unveil the structure and dynamics of the ice surface [6, 7]. We show that in the range from -80 to 0 C, the main crystal facets of ice undergo a sequence of alternating structural surface phase transitions that result in the anomalous increase of step free energies and the crossover of crystal growth rates exactly as required to explain the Nakaya diagram [7].

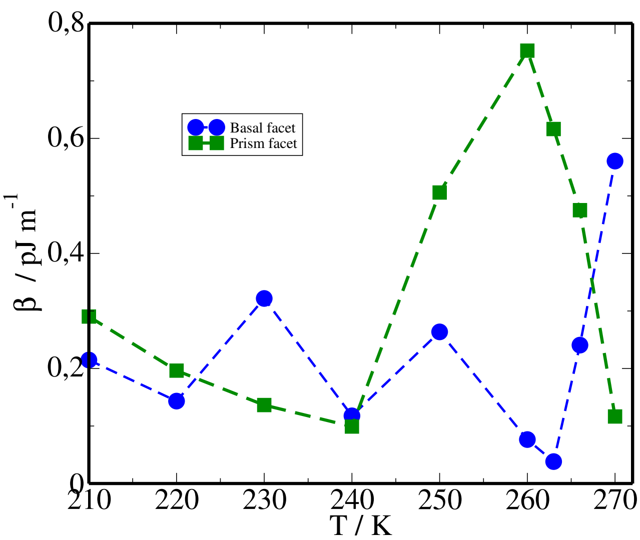

Fig. 1. Step free energies of ice below a premelting film. Results are shown for basal and prism facets as a function of temperature as calculated from the spectrum of surface fluctuations of the TIP4P/Ice water model. The free energies crossover twice, resulting in a crossover of crystal growth rates and the alternation of crystal habits from plate to column like hexagonal prisms twice.

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