Molecular mechanism of heterogeneous ice nucleation in the atmosphere

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Abstract:

Ice nucleation in the atmosphere has a central role in precipitation formation, the hydrological cycle, and Earth's radiation budget. This process occurs mainly on ice-nucleating particles (INPs), which provide surfaces that facilitate ice formation, rather than in pure supercooled water. Potassium (K) feldspar, a widespread aluminosilicate mineral, is recognized as the most efficient INP [1], and previous studies have attributed its high ice nucleation activity to the exposure of (100) surfaces at structural defects [2] (see Fig. 1). In this talk, I will discuss our recent molecular dynamics (MD) simulations that overturn the established mechanism for this phenomenon and lead to a new molecular-level picture of how ice forms in the atmosphere. I will first describe how we trained a highly-accurate machine-learning (ML) model for the atomic interactions in this system, based on electronic-structure calculations. Then, I will show the application of this ML model to investigate ice nucleation on all possible K-feldspar surfaces. We identify the (110) surface, exposed at defects such as steps, as the most active plane for ice formation, challenging previous evidence that pointed to the (100) surface as the most relevant one. The (110) surface uniquely structures interfacial water into an arrangement resembling the order shown by liquid water on the (110) surface of cubic ice, providing thus an optimal template for nucleation (see Fig. 2). Using advanced sampling, a methodology aimed at simulating rare events, we directly observe the dynamics of the formation of ice clusters on a K-feldspar surface for the first time. We show that, in the initial stages of this process, the ice clusters exhibit a structure compatible with cubic ice, rather than the more stable hexagonal ice polymorph found in macroscopic ice crystals and underpinning the characteristic shape of snowflakes. Moreover, we demonstrate that the orientation of these nascent ice clusters fully agrees with the available experimental evidence (see Fig. 3). Taken together, these insights provide a comprehensive explanation for the very high ice nucleation efficiency of K-feldspar, and represent the most detailed characterization to date of the mechanism for ice formation in our planet's atmosphere.

References

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[2] A. Kiselev et al., Science, 355(6323) (2017) 367–371.
[3] P. M. Piaggi et al., Faraday Discuss. 249 (2024) 98–113
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Figures

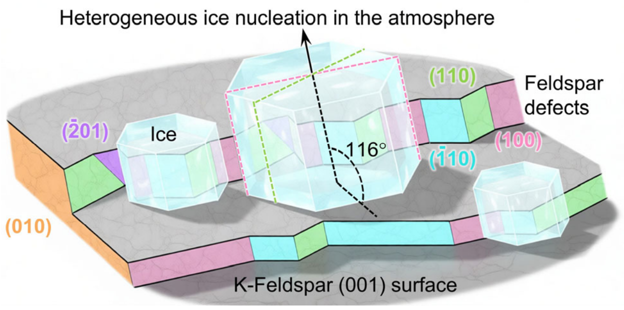

Figure 1. Schematic illustration of the heterogeneous ice nucleation at K-feldspar defects with exposed non-perfect cleavage planes. The pink and green dashed lines highlight the first and second prismatic surfaces of ice, which are approximately parallel to the (100) and (110) planes of feldspar, respectively.

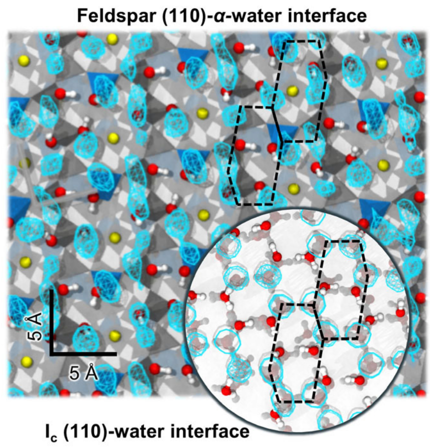

Figure 2. Water-density isosurfaces at 70 molecules/nm3 in the x–y plane at the feldspar (110)-α-water interface and the cubic ice, Ic, (110)-water interface.

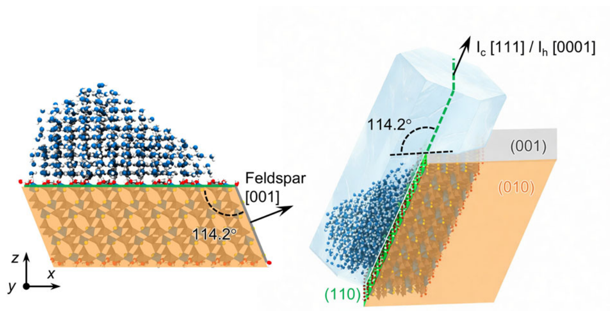

Figure 3. Orientation and lattice of ice formed on the K-feldspar (110)-α surface (left panel). Illustration of the ice basal-plane axis orientation relative to the feldspar (001), (010), and (110) planes (right panel). Note the basal (0001) plane of hexagonal ice (I_h) is equivalent to the (111) plane of cubic ice (I_c).