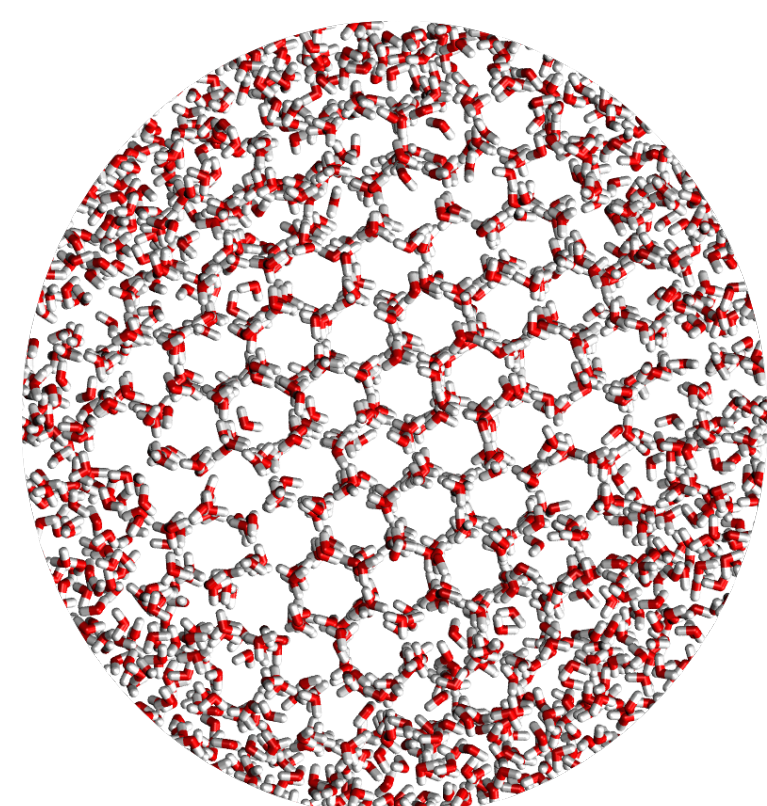


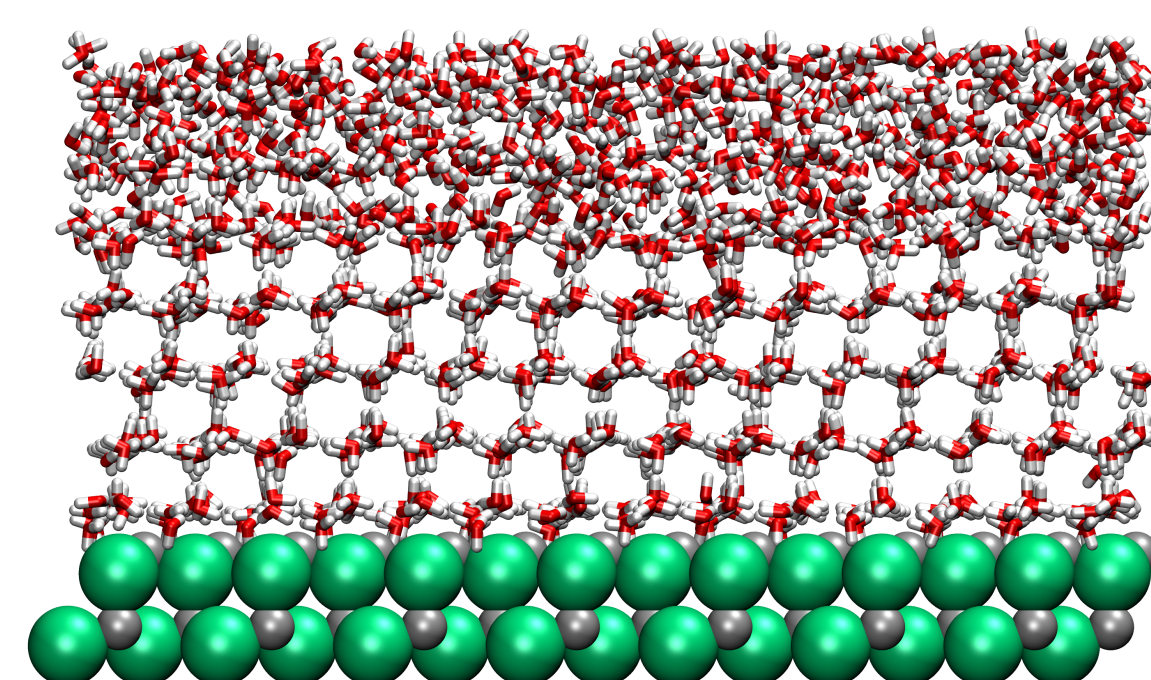
Introduction

Ice nucleation

The crystallization of ice from liquid water occurs both homogeneously and heterogeneously. Most atmospheric ice nucleation occurs heterogeneously, at the surface of atmospheric aerosols. These atmospheric aerosols can include organic compounds, some bacteria, mineral dusts, pollens, and soot particles. While homogeneous ice nucleation requires temperatures as low as -37°C , heterogeneous ice nucleation can occur at much higher temperatures.



Homogeneous ice nucleation

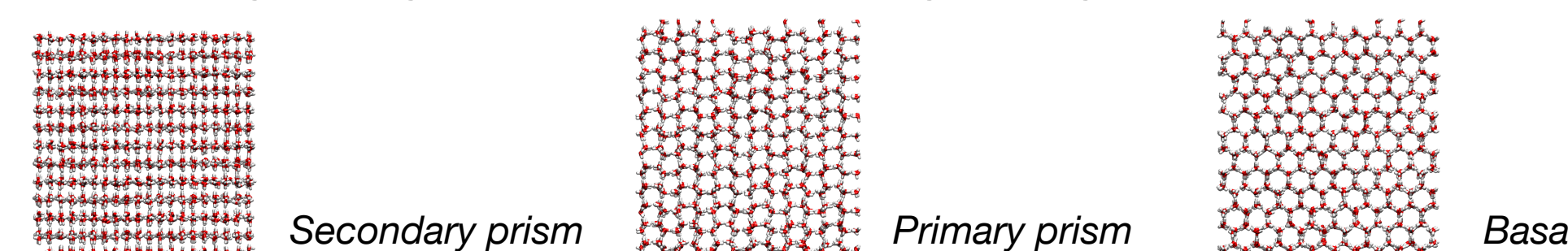


Heterogeneous ice nucleation on a silver iodide surface

While experiments have been successful in both identifying active ice nucleating agents and determining the temperatures at which they can cause the onset of freezing, it remains difficult to predict whether a surface will be an ice nucleating agent and equally challenging to design surfaces with ice nucleating ability. Knowledge of how these surfaces template the formation of ice on the molecular level would greatly enhance our ability to design surfaces with hyperactive ice nucleating ability.

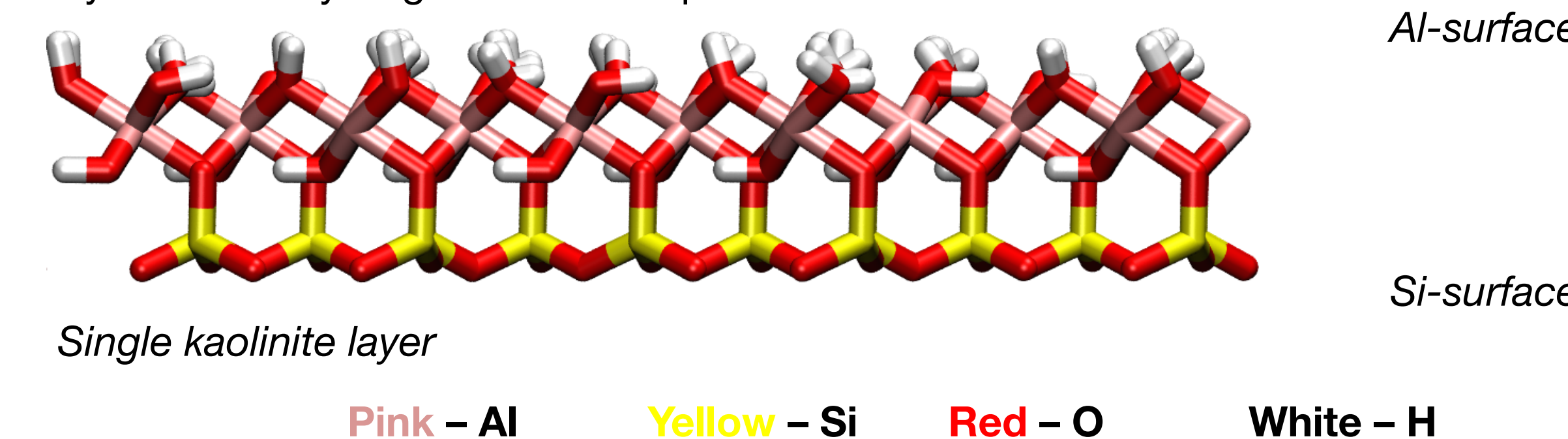
Faces of ice I_h

We consider three commonly growing faces of the ice I_h crystal: primary prism, secondary prism (fastest growing), and basal (slowest growing).



Surfaces of interest

Silver iodide (AgI) is known to be an excellent promoter of ice nucleation. Its ability to cause freezing at temperatures as high as -4°C and relative safety have made its industrial use important as a cloud seed. Kaolinite is commonly found in atmospheric mineral dusts and is known to nucleate ice at temperatures as warm as -10°C . It is a layered mineral, with each layer consisting of repeating alumina tetrahedra stacked atop repeating silica tetrahedra. Each layer is neutrally charged. The aluminum surface of the kaolinite layer is hydroxylated and has the ability to act as both a hydrogen bond donor and acceptor. The silicon surface only acts as a hydrogen bond acceptor.



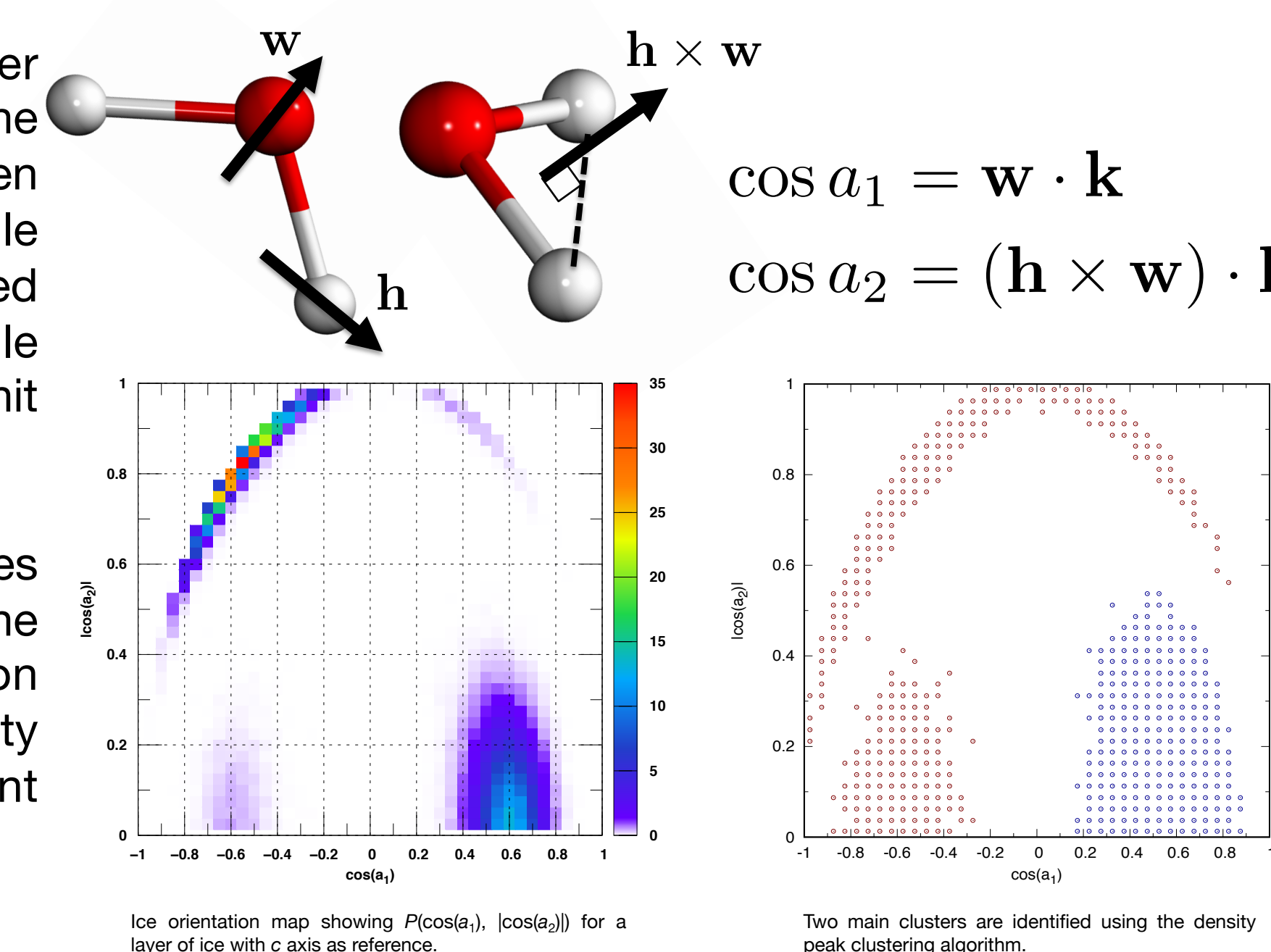
Methods

Molecular dynamics simulations

Molecular dynamics (MD) simulations allow us to study the motions of molecules in atomistic detail. We use MD to simulate the behavior of water molecules at surfaces. In these simulations, AgI is modeled using a potential developed by Hale and Kiefer, kaolinite is modeled using the CLAYFF force field, and water is represented using the TIP4P/Ice water model. All surface atoms are kept fixed, except for interfacial hydrogen atoms of the kaolinite surface which are allowed to move.

Ice orientation map

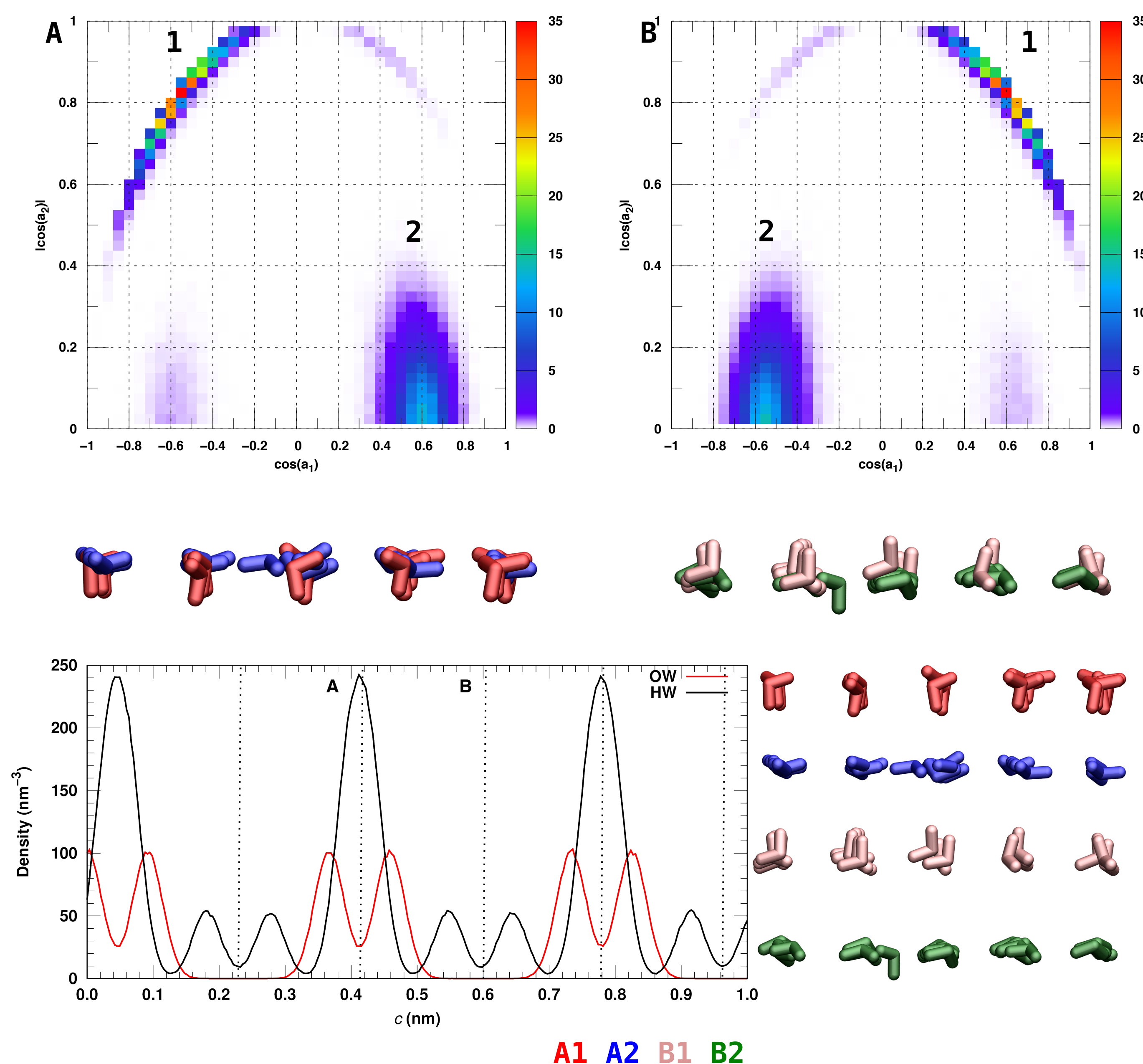
To identify the predominant orientations of water molecules in ice with respect to specific axes, we define two angles, a_1 and a_2 . a_1 is defined as the angle between the unit vector in the direction of the water molecule bisector, \mathbf{w} , and a reference unit vector, \mathbf{k} . a_2 is defined as the angle between the plane of the water molecule with unit normal vector $\mathbf{h} \times \mathbf{w}$ and the reference unit vector.



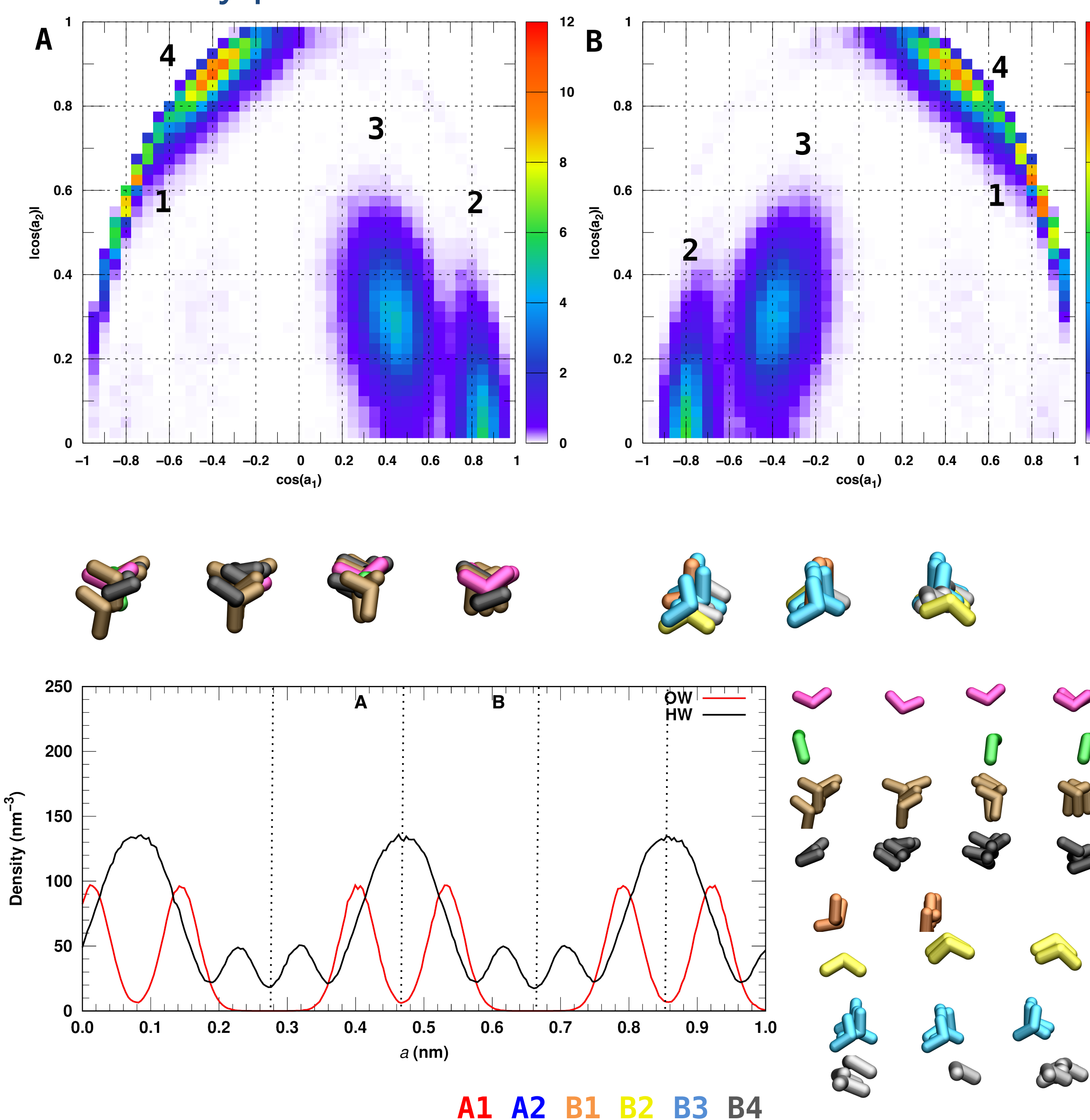
Results

Orientational signatures of ice faces

Basal face

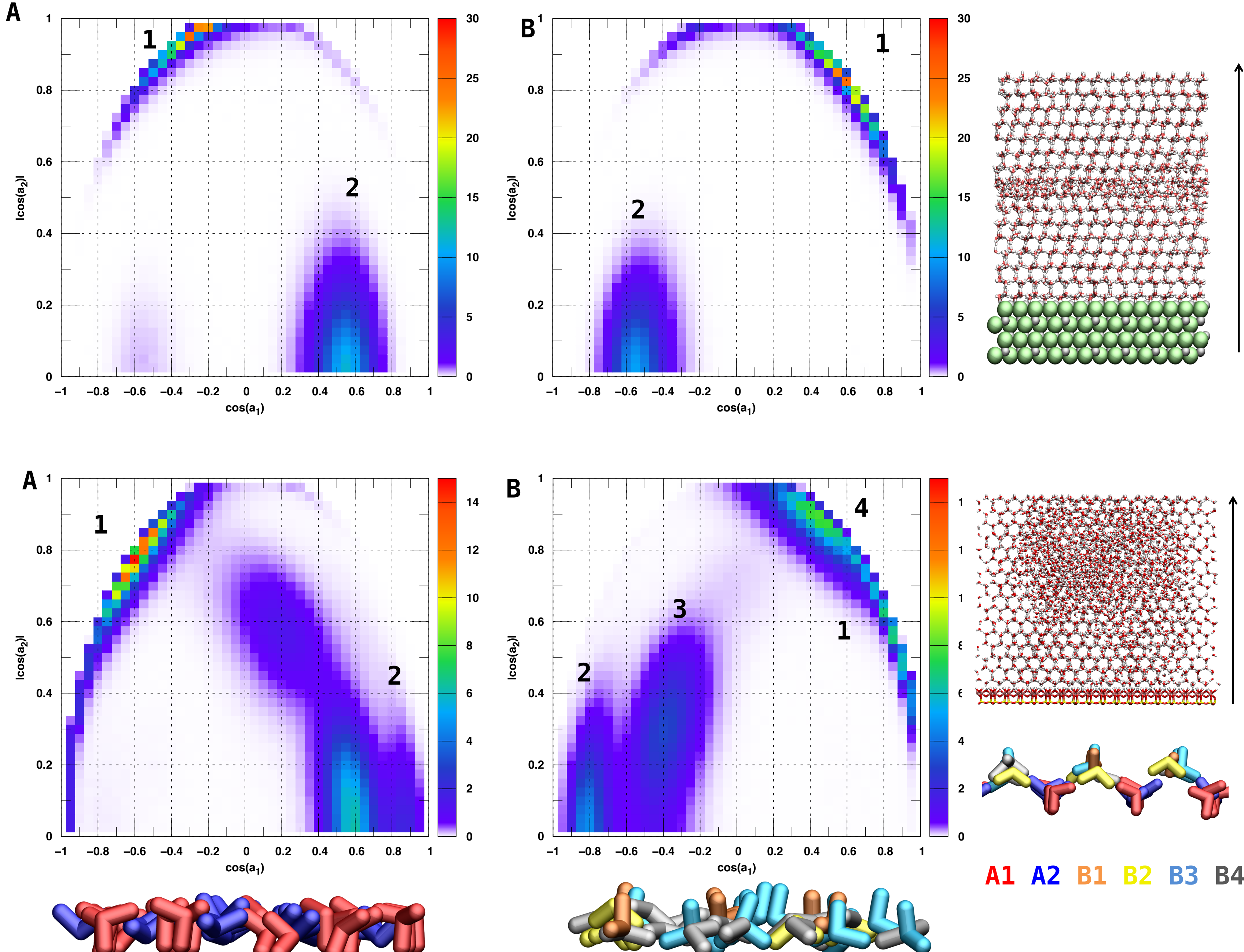


Secondary prism face

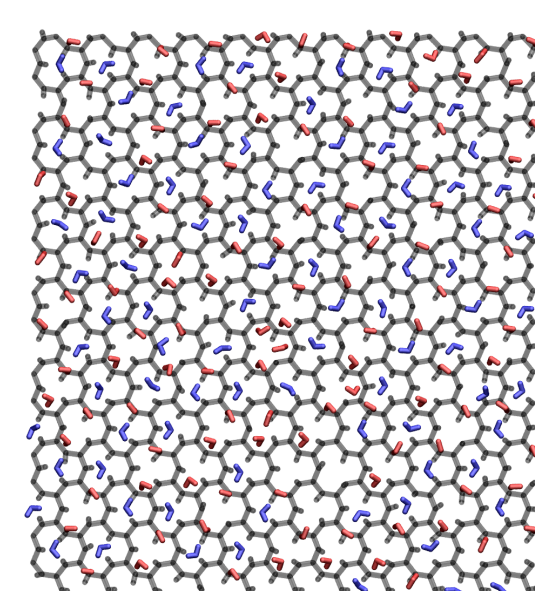
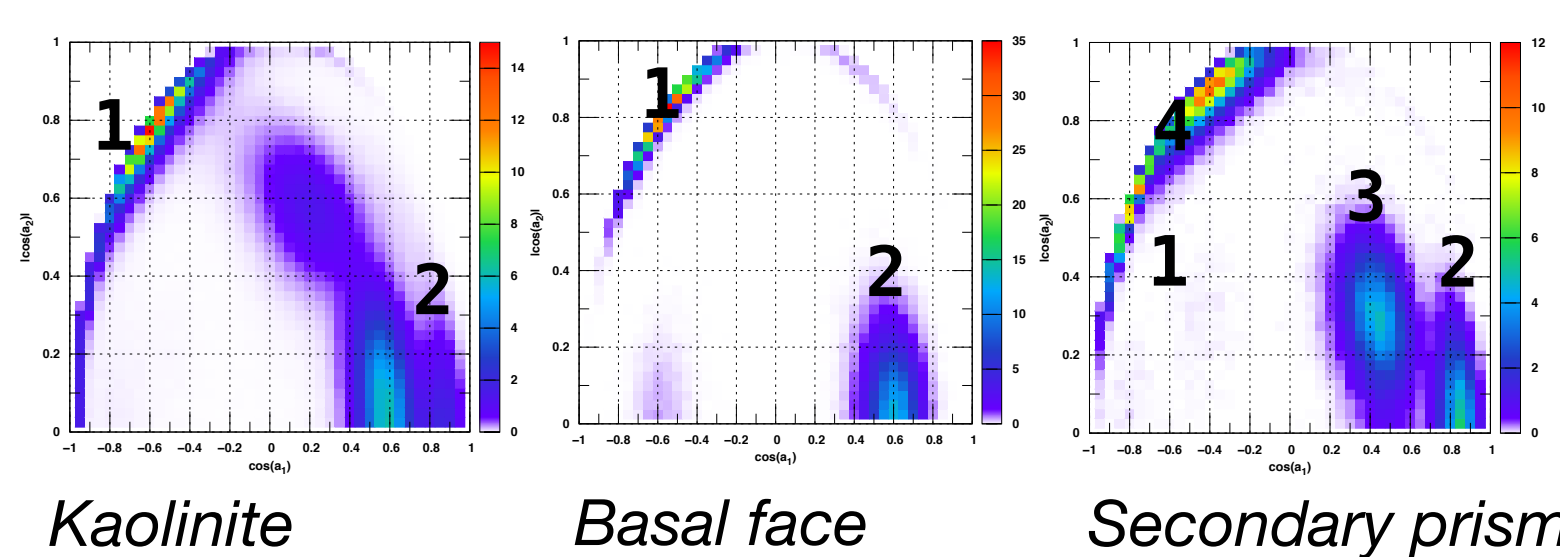


Application to heterogeneous nucleation

Silver iodide



Conclusions and Future Directions



What causes this layer to template the secondary prism face?

The first layer of ice grown on kaolinite is orientationally intermediate between the basal face and secondary prism face.

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Acknowledgements

PI: Prof. Yu-Shan Lin
Sean McHugh
Dan McKeen
Arya Mekkat
Diana Slough
Dr. Hongtao Yu

Tufts Summer Scholars Program
Dr. Anne Moore
Ashley Wilcox

