Ice Nucleation in Aerosols Containing Biomolecules

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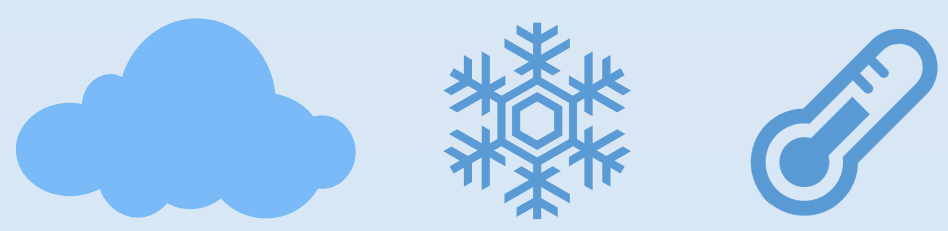
1. Why Study Ice Nucleation in Aerosols? [1]



The nucleation of ice in droplets is important for understanding:

1. Weather and meteorological models
2. Industrial processes such as flash freezing
3. The radiative forcing of clouds, and thus, climate models

Despite these applications, heterogeneous ice nucleation is still poorly understood, especially with respect to which properties of a material give it good nucleating ability.

2. Proposed Research Strategy

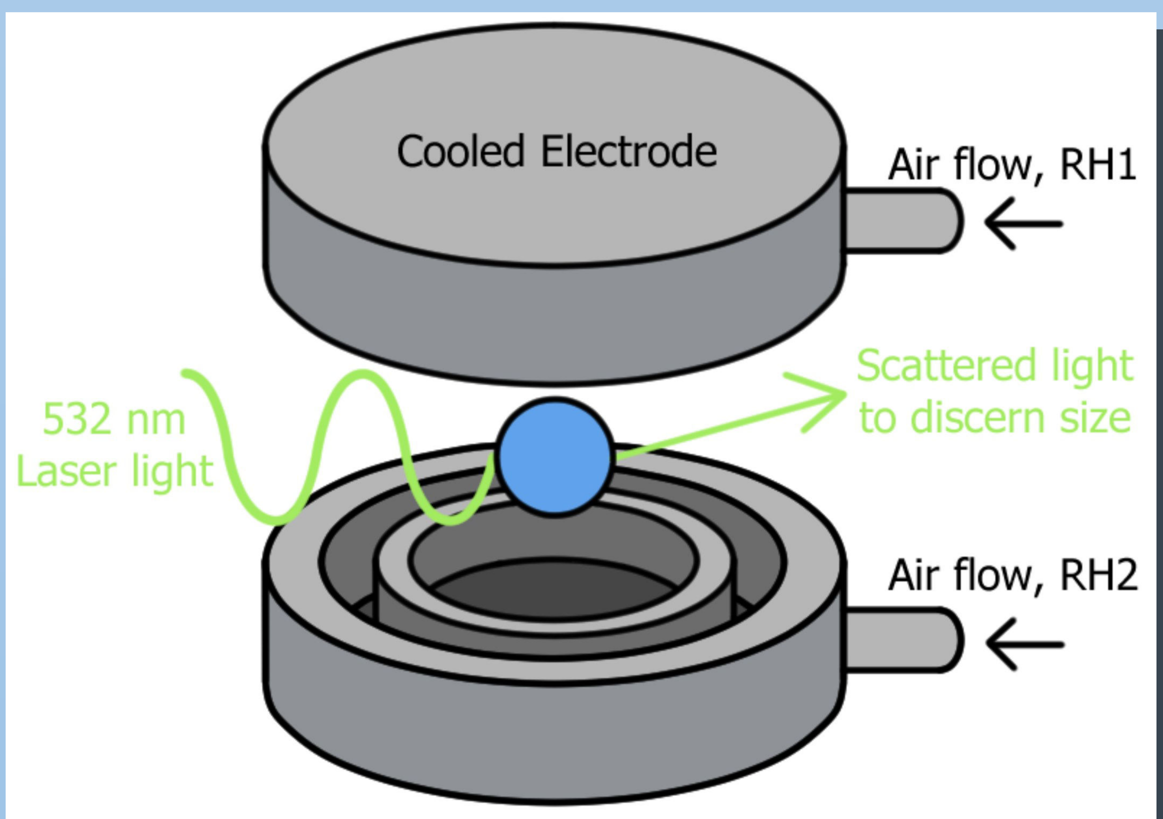
1. Construct an **electrodynamic balance (EDB)** suitable for examining the freezing behaviour of levitated single aerosol droplets. This should be able to explore micron scaled droplets

2. Using the EDB apparatus, explore the freezing behaviour of droplets containing the protein apoferritin/ferritin under different pre-treatment conditions: heat treatment, pH and concentration

3. Begin exploring droplets containing surfactants. Investigate different surfactant concentrations, droplet sizes and surfactants of different dimensions.

4. Continue with freezing experiments of droplets containing DNA origami sheets organised into wedges. By varying the angle between the sheets, explore substrate effects. Also conduct experiments with unannealed DNA.

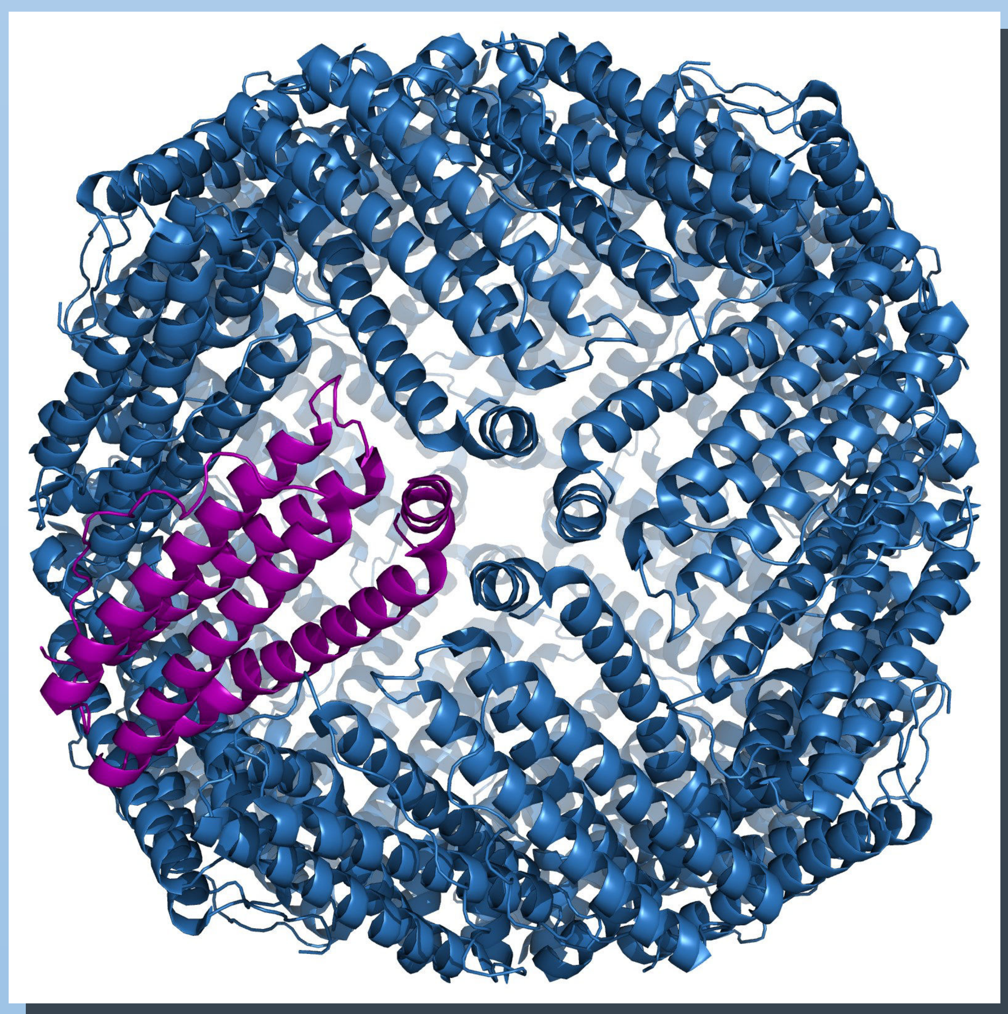
3. The EDB Design (TOP LEFT) [2]



The EDB uses a combination of AC and DC electric fields to levitate a single charged droplet in place. Using scattered light from a laser, the size of the suspended droplet can be found.

Conditions in the device can be varied using attached coolers/heaters and through running different humidity gases through the chamber.

The EDB benefits from droplets 100 to 1000 times smaller than droplet array methods leading to less contaminant driven nucleation.

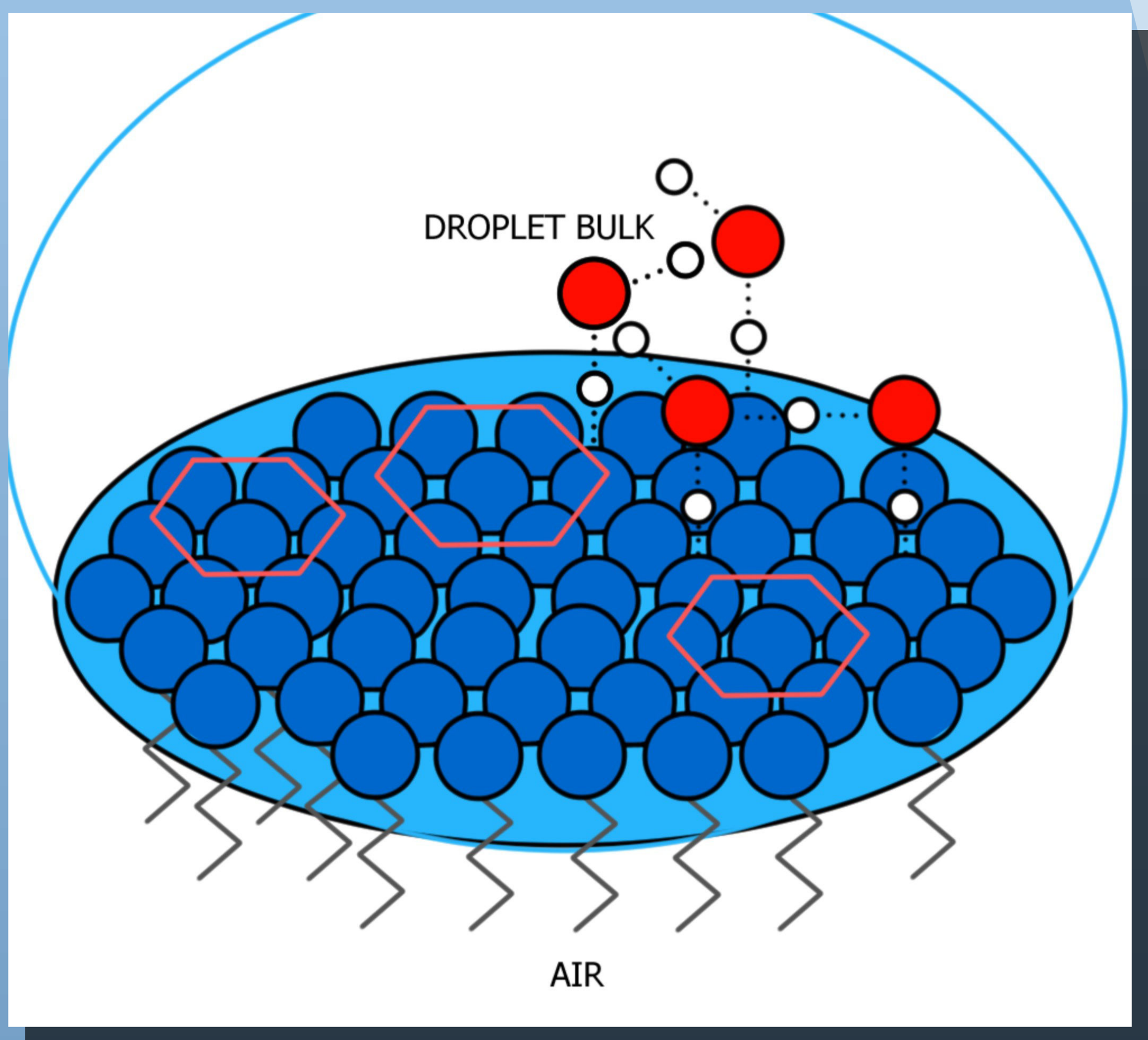


4. Apoferritin (TOP RIGHT) [3]

Apoferritin is a protein that stores iron. Despite this original evolutionary purpose, the protein is an exceptional ice nucleator.

The protein is believed to primarily nucleate ice growth through forming larger protein aggregates. Smaller droplets from the EDB provide a good environment to explore the effect of these rare structures.

The protein has a well studied structural behaviour under denaturing conditions such as at low pH and heat treatment. Exploring the ice nucleating ability of the protein under these conditions provides better insight into the biomolecule structures that best nucleate ice.

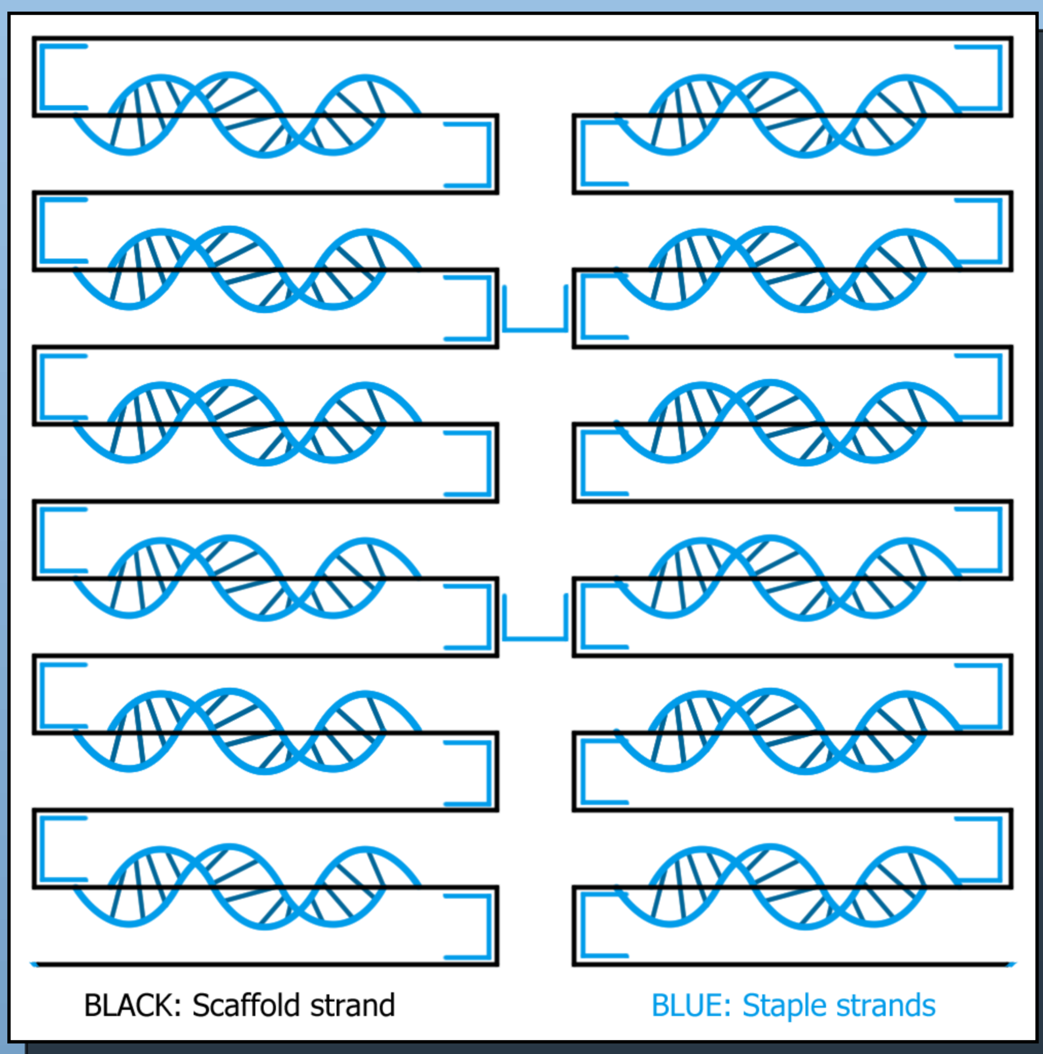


4. Surfactants [4]

Surfactant molecules in a droplet will generally accumulate at the air-water interface ie. The surface of the droplet.

Hydrophilic surfactant molecule heads (circles) pack in a hexagonal arrangement, similar to that of ice's crystal structure.

Different surfactant concentrations and molecules can be explored to see how parameters such as lattice matching effect the "scaffolding" of the ice and thus affect the nucleation rate.



5. DNA origami [4]

DNA by itself is a known ice nucleator but this ability can be augmented by changing the shape the DNA conforms to. Using a combination of long tailored DNA "scaffold" strands and shorter "staple" molecules, rigid 2D and 3D structures can be formed out of sheets of DNA.

By combining two sheets of this material, a wedge can be formed with a tuneable pitch angle.

Similar to the planar case from surfactants, the concave pit of the wedge can act to change the volume of the critical ice embryo, changing the rate of ice nucleation.

In these experiments, the angle between wedges can be varied, and this substrate effect can be explored.

Previous computational work has explored similar (though much smaller in scale) wedges formed from graphene and showed a large dependence between the interior angle and the rate of nucleation. [5]

References

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2. Stockel, T. (2005). Rates of Homogeneous Ice Nucleation in Levitated H2O and D2O Droplets. The Journal of Physical Chemistry A, 109(11), 2540-2546.
3. Kim, M. (2011). pH-Dependent Structures of Ferritin and Apoferritin in Solution: Disassembly and Reassembly. Biomacromolecules, 12(5), 1629-1640.
4. Jamieson, S. (2005). First Study on the Effects of Interfacial Curvature and Additive Interfacial Density on Heterogeneous Nucleation. Ice Crystallization in Oil-in-Water Emulsions and Nanoemulsions with Added 1-Heptacosanol. *Crystal Growth & Design, 5(2), 451-459.*
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