# Development and preliminary results of the low-temperature gas evaporation method to understand nucleation conditions of water ice and related materials

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In our previous studies, we have shown that the key factors governing nucleation from the gas phase are the melting point depression of nanoparticles and sticking probability (dimer formation) [1,2]. In long years, nucleation theory has been known to be completely unable to explain the results of experiments and molecular dynamics simulations. In contrast, we have clarified that the latest nucleation theory can explain experimental results by using appropriately determined sticking probabilities and surface free energies of nanoparticles as physical parameters [1], and by including chemical potential differences that consider the nucleation pathway [2].

To understand the nucleation processes of ice, we developed a low-temperature gas evaporation method to determine the sticking probability and surface free energy of nanoparticles of ice. The gas evaporation method has been used to make nanoparticles of refractory materials through homogeneous nucleation during the cooling process of high-temperature vapor generated by heating and evaporating the starting material [3]. By applying interferometry to this method, two physical quantities (surface free energy and sticking probability), which are essential for understanding the nucleation process, can be obtained. In addition, infrared spectral measurements can reveal the nucleation pathways.

The low-temperature nucleation chamber has a double-tube structure. The inner chamber, where ice nucleation takes place, is filled with neon gas to reduce the mean free path and cooled by liquid nitrogen. The evaporation source was also cooled with liquid nitrogen. The space between the inner and outer chambers is evacuated to thermally insulate them and prevent condensation.

Water vapor generated by heating and sublimating ice in the low-temperature nucleation chamber is cooled by neon gas, and after becoming supersaturated, it undergoes homogeneous nucleation to form nanoparticles. This process was observed in situ using a double-wavelength Mach-Zehnder laser interferometer and an infrared spectral system. The interferometer can quantitatively determine the temperature and concentration fields from evaporation to nucleation. The infrared system can investigate the changes in the crystal structure of water and ice particles during the nucleation process.

In the experiment, the interferometer succeeded in capturing the moment of nucleation, and the infrared system also succeeded in capturing the temporal change in absorption intensity. This means that the nucleation process of ice nanoparticles from water vapor was captured. This achievement is expected to lead to the construction of a nucleation model that can predict the state of ice dust formation that repeatedly sublimated and condensed in the primordial solar nebula 4.6 billion years ago.

## References

[1] Y. Kimura, K. K. Tanaka et al., 2017, Science Advances 3, e1601992.
[2] S. Ishizuka, Y. Kimura et al., 2018, Nature Communications 9, 3820.
[3] C. Kaito, 1978, Japanese Journal of Applied Physics 17, 601.