Effects of deposition temperature on structure of amorphous ice

*Yui Ono[1], Kengo Oki, Tomoko Fukazawa[1,2]

1. Meiji University, 2. The Japanese Society of Snow and ice

In interstellar molecular clouds, elements such as hydrogen, oxygen, carbon, and nitrogen deposit on dust grains, and forms various molecules such as H_2O, CO_2, NH_3, CH_4, H_2CO, CH_3OH, and so on. The deposited water exists as amorphous ice. Because molecules undergo chemical evolutions through various processes on amorphous ice, amorphous ice has an important role for the chemical evolutions in the universe. Amorphous ice formed from a vapor deposition is principally classified into two types: low-density amorphous (LDA) and high-density amorphous (HDA) ices. HDA ice is transformed into LDA ice with warming [1]. However, the mechanism of the phase transition is less conclusive, because the transition temperature depends on experiments [2]. To investigate the effects of thermal history on structure of amorphous ice, a dependence of deposition temperature on infrared spectrum of amorphous ice was analyzed.

Amorphous ice was prepared with vapor deposition of distilled and degassed water on a substrate of oxygen-free copper at a temperature of 43.5–120.5 K. The total pressure in the vacuum chamber was kept at about 5.0×10^{-5} Pa during the deposition. After the deposition of amorphous ice, the substrate was cooled to 43.5 K at a rate of 2 K/min. Then, the sample was warmed to 176 K at a rate of 1–4 K/min. The IR spectra were measured using Shimadzu IRPrestage-21 at every 15 seconds during deposition, and measured at 2 K intervals during cooling and warming.

The effects of the deposition temperature on the structure of amorphous ice was analyzed using the spectra at 43.5 K after cooling from their deposited temperatures. The result shows that the wave number of the O–H symmetric stretching mode depends on the deposition temperature. The wave number gradually decreases as the deposition temperature increases at temperature below 107 K. The decreasing rate changes at 63 and 76 K. This indicates that the HDA ice is transformed into LDA ice via an intermediate structure in the temperature range of 63–76 K. Furthermore, a significant decrease in the wave number was observed at 108 K. This decrease is associated with the transition from the LDA ice to crystalline cubic ice (Ic). The transition point is lower than that observed in the warming process from the HDA ice (i.e., 145–160 K [1]). The lower shift in the transition point is caused by a roughness of its surface. A liquid-like structure in a rough surface can be a cause of the lower shift of the transition point, because the rearrangement motion of water molecules is enhanced with an increase in amplitudes of the thermal vibrations [3].

References

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