Development of high-pressure RHEED and IR spectroscopy for understanding the structure of vapor-deposited ice under terrestrial atmospheric conditions

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Water (H_2O) ice has many forms. Twenty-one different crystalline structures have been observed, named ices I-XIX. Water is ubiquitous in the universe. In the extra-terrestrial space, it is an abundant intersteller molecule. Ice covers the surface of dust grains in molecular clouds, which are cold (10 K) and dense (10^4 molecules cm^{-3}; 10^{-12} Pa) interstellar regions. Water ice also exists in the Earth's atmosphere [1]. Polar mesospheric clouds, also known as noctilucent clouds, are the highest clouds in Earth's atmosphere, which occur at the heights of about 82 km at temperature below about 145 K under partial vapour pressure of 10^{-7}–10^{-5} Pa. The formation of polar mesospheric clouds is a topic of intense research because they are beneficial to understanding the physics and chemistry of the mesosphere and lower thermosphere and also serve as potential indicators of global climate change.

Recently, we developed an experimental apparatus for the in situ structural analysis of vapour-deposited ice through a combination of reflection high energy electron diffraction (RHEED) and infrared reflection–absorption spectroscopy (IRRAS), and reported that the structures of the growing ice under polar mesospheric temperature (120 K) and vapour pressure (10^{-6} Pa) conditions differ depending on the ice size (thickness), that is, the ice appeared to grow in three steps during vapour deposition, being amorphous water for the first 15 nm, then cubic ice Ic up to 50 nm, and finally hexagonal ice Ih subsequently [2]. Although the current RHEED-IRRAS apparatus powerfully works to study the structure of vapour-deposited ice, it requires a high vacuum condition to protect the turbo molecular pump in the vacuum chamber and the electron gun for RHEED from damaging. Thus, the current apparatus cannot simulate the total pressure in the mesosphere (about 1 Pa). In addition, it is impossible to study the ice structures in other Earth's clouds, such as polar stratospheric clouds, important factor of ozone depletion, which form at below 188 K at heights of 17–31 km under partial vapour pressures of 10^{-3}–10^{-2} Pa with the total pressures of 10^3–10^4 Pa [3,4,5,6].

For understanding the structure (amorphous water, cubic ice Ic or hexagonal ice Ih) of ice in the Earth's clouds, we are now constructing a new apparatus to study the ice structures under higher pressure conditions using a combination of high-pressure RHEED and IR spectroscopy. Two-stage differential pumping system is employed to protect the filament of the gun from burning out even when the pressure of the sample chamber reaches 10 Pa. The distance between the electronic gun and the screen is designed to be shorter than the mean free path of electrons under 10 Pa for RHEED measurements. A silicon (Si) substrate is used for RHEED and IR measurements. The substrate can be cooled down to 77 K using liquid nitrogen, and heated by a heater up to 230 K, at which the vapour pressure of ice Ih reaches 10 Pa [5]. Our new apparatus will be able to simulate polar mesospheric conditions including its total pressure (1 Pa). Simulating the partial vapour pressure and temperature of polar stratospheric conditions is also possible for studying the ice structures of polar stratospheric clouds.

References

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