

Low temperature crystallization of free-flying silicate nanoparticles investigated by in-situ IR measurement experiment

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Dust is typically 100 nm sized nanoparticles which can be observed ubiquitously in the universe. Dust forms from the high temperature gas in the out flow of evolved stars and dispersed into interstellar space. Silicate dust is one of the most abundant minerals in the universe including, shells around evolved stars [1], disks around young stars [2], comets [3] and so on. So its formation mechanism is the key process to understand the lifecycle of dust. Especially, 10 μm IR band structure from 8 μm to 12.5 μm in wavelength arising from Si-O stretching provides us mineralogical character of silicate. The Infrared Space Observatory mission revealed the existence of crystalline silicates around evolved stars based on the 10 μm band feature mainly attributed to amorphous silicate [4]. Numerous laboratory experiments to reproduce the observed spectra such as direct condensation [e.g. 5] and annealing of amorphous silicates [e.g. 6] showed variation in the IR spectra due to structure, chemical composition, temperature, size and shape, and proposed formation mechanisms of crystalline silicates. Nevertheless the scenario is not fully understood. One of the most important discrepancies concerning the dust formation process is a detection of an IR feature attributed to crystalline silicates at low temperature region, typically <300 K [1] in contrast to amorphous silicates at high temperature region [4]. Low temperature crystalline silicates cannot be explained by direct condensation or annealing involving high temperature process.

Recently, we have investigated new IR measurement technique for free-flying nanoparticles which enabled direct comparison with astronomical observation without KBr medium effects which pervert its band structure such as peak wavelength, FWHM and relative intensity [7]. Applying the new IR technique, we investigated condensation of Mg-bearing silicate from thermally evaporated magnesium and silicon oxide under the atmosphere of O₂ and Ar based on 10 μm band.

In-situ IR measurement revealed initial condensates were amorphous or droplet of Mg-bearing silicate and its crystallization took place at <500 K. Furthermore, crystallization kept proceeding through lower temperature region. Produced particles showed core-mantle like structure, amorphous silica covered with polycrystalline forsterite observed by Transmission Electron Microscope.

Prevailing annealing experiments reported that 1000 K is required for crystallization of forsterite [8]. This critical discrepancy may be explained by nano size effects. When immoderately small particle nucleates, a particle takes metastable amorphous or droplet phase because of lower melting point of a nanoparticle [9] and larger diffusion coefficient of molecules in a nanoparticle distinct from in bulk [10]. In case the condensates were droplet due to the size effects, activation energy of crystallization is significantly low compared to amorphous [11]. We concluded such characteristic phenomena in nanometer scale enabled low temperature crystallization in the same way as the circumstellar environments.

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