

Synchrotron IR Microscopy of Manually Arranged Aggregate Particles

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Cosmic dust grains are in most cases not well segregated mutually in nature but rather are formed in the state of aggregates, which strongly influences the optical properties. The aggregation effects such as degree of porousness, size and shape of aggregates on infrared (IR) band profiles are not well understood in detail because the aggregate structures are not sufficiently well defined. The overall morphological effect (considering size, shape, and agglomeration state all together) on IR spectra has been verified by aerosol spectroscopy. However, the particles, which were produced by an aerosol generator, were hardly separated from each other. Thus it was virtually impossible to quantify the relative importance of each morphological effect.

In order to understand the influence of particle aggregation on the diagnostic phonon bands of solid particles, we carried out IR microscopy of well-defined aggregates which were systematically manufactured on a highly transparent substrate (Si) in mid-IR region by means of a scanning electron microscope (SEM) coupled to a focused ion beam (FIB) at Max Planck Institute for the Science of Light (Germany). The positioning of particles was achieved by a nanomanipulator, which was controlled under the SEM, enabled the defined motion of a probe by the extension or contraction of piezo crystals under electrical voltage. Six complex aggregate structures (a single particle, linear 2, 3, 4 particles, a triangle shape with 3 particles, a diamond shape with 4 particles) were all composed of 1 μ m-sized amorphous SiO₂ monosphere particles. A clear transition of the Si-O stretching vibration band at 10 μ m (absorption intensity, band broadening, peak shift), which depends on the numbers of particles as well as the structure of them, is anticipated to observe via the mid-IR extinction measurements. The IR extinction measurements have been performed at SOLEIL synchrotron facility in Paris, France, since IR light from the synchrotron can be focused to sample spots close to the diffraction limit which allows achieving high spatial resolution and is possible to improve signal-to-noise ratio. In addition, a polarizer was employed for the extinction measurements so as to investigate the polarization effects caused by the particle orientation except for the single particle.

Extinction spectra obtained by various patterns of particle arrangements can directly compare not only with observed spectra, but also serve as a benchmark for corroborating the precision of the light absorption/scattering theoretical calculations such as Mie theory, discrete dipole approximation (DDA), and finite-difference time-domain (FDTD) method.

We report herein the first detailed experimental results obtained by synchrotron IR microscopy of systematically arranged 1 μ m-sized SiO₂ monosphere particles considering the polarization effects caused by the particle orientation. Furthermore, the experimentally measured extinction spectra were compared with theoretical calculations.