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Inhomogeneities in particle composition of single, levitated aerosol particles observed by Mie resonance spectroscopy

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Recent observations have indicated that organic aerosol particles in the atmosphere may exist in an amorphous semi-solid or even solid (i.e. glassy) state, e.g. [1]. The influence of highly viscous and glassy states on the timescale of aerosol particle equilibration with respect to water vapor have been investigated for some model systems of atmospheric aerosol, e.g. [2,3]. In particular, it has been shown that the kinetics of the water absorption/desorption process is controlled entirely by liquid-phase diffusion of water molecules for a highly viscous aerosol particle. A liquid phase diffusion model based on numerically solving the non-linear diffusion equation predicts strong internal gradients in water concentration when condensed phase diffusion impedes the water uptake from the gas phase [2]. Here we observe and quantify the internal concentration gradients in single, levitated, micron size aerosol particles of aqueous shikimic acid using elastic Mie resonance spectroscopy.

A single, aqueous particle is levitated in an electro-dynamic balance (for details see [2]), dried for several days at room temperature, cooled to the target temperature and exposed to a rapid change in relative humidity. In addition to measuring the elastically backscattered light of a “white light” LED source and recording the full spectrum with a spectrograph as in [2], we use a tunable diode laser (TDL) to scan high resolution TE- and TM spectra. This combination allows observing various Mie resonance mode orders simultaneously. Since we perform the experiment at low temperatures and low humidities the changes in the Mie-spectra due to water uptake are sufficiently slow to resolve the kinetics.

Experimental Mie resonance spectra are inverted to concentration profiles of water within the particle by applying the numerical diffusion model [2] in conjunction with Mie calculations of multilayered spheres [4].

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