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Mie resonance shifts as experimental mean to track water diffusion fronts inside of a highly viscous aerosol particle

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Field measurements indicate that atmospheric secondary aerosol particles (SOA) can be present in a highly viscous, glassy state [1]. In contrast to particles in a liquid state, the gas phase equilibration is kinetically limited and governed by condensed phase diffusion. In recent water diffusion experiments on highly viscous single aerosol particles levitated in an electrodynamic balance (EDB) we observed a characteristic shift behavior of the Mie whispering gallery modes (WGM) indicative of the changing inner radial structure of the particle, thus providing us with an experimental method to track the diffusion process inside the particle.
A micron-sized shikimic acid droplet is dried in the EDB over several days at room temperature and then cooled, forming a homogeneous semi-solid particle. When such a particle is exposed to an abrupt increase in relative humidity, the rapid gas phase diffusion and strong concentration dependence of the diffusion coefficient in the condensed phase lead to extremely steep water concentration gradients inside the particle, reminiscent of diffusion fronts. The resulting quasi step-like concentration profile motivates the introduction of a simple core-shell model describing the morphology of the non-equilibrium particle during humidification. Using high resolution Mie resonance spectroscopy, the subsequent particle growth and reduction of the shell refractive index can be observed as red- and blueshifts of the WGM, respectively. Modes of the same order will simultaneously undergo a maximal blueshift and the time at which the maximal blueshift occurs increases with increasing mode order. This can be linked to the radial energy distribution of the modes in the particle, as higher mode orders tend to penetrate deeper into the particle. The individual WGM blueshifts are attributed to a certain core-shell radius ratio derived from model calculations [2]. If supplemented with growth information obtained from the WGM redshift and thermodynamic equilibrium data, we can infer a comprehensive picture of the time evolution of the diffusion fronts in the framework of our core-shell model. The measured time dependent concentration profile is then compared with simulations solving the non-linear diffusion equation [3].
[1] Virtanen, A., et al., Nature, 467, 824-827, 2010
[2] Kaiser, T., Schweiger, G., Computers in Physics, Vol. 7, No. 6, 682-686, Nov/Dec 1993
[3] Zobrist, B., Soonsin, V., Luo, B.P., Peter, T. et al., Phys . Chem. Chem. Phys., 13,3514-3526, 2011