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Phase transition dynamics of single optically trapped aqueous potassium carbonate particles†

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1 Bulk sample Raman scattering measurements

Single aqueous K_2CO_3 particles are trapped with a counter propagating tweezer and dried while being interrogated with time resolved broad band light scattering (BLS) and Raman scattering measurements. Particle size and refractive index (Cauchy parameters, $m_0(\lambda)$, m_1) are determined with BLS measurements. The Raman shift is used to monitor the CO_3^{2-} symmetric stretching peak position, $\nu_1(CO_3^{2-})$. See the main text for the experimental details.† Measured values of $\nu_1(CO_3^{2-})$ are compared with bulk solution measurements in order to determine the particle concentration prior to supersaturation. The bulk aqueous K_2CO_3 solution saturation point is 8.1±0.7 mol kg^{-1} corresponding to 43% relative humidity (RH) in equilibrium.

Table 1 Bulk solution measurements.

Molality/ mol kg^{-1}	$\nu_1(CO_3^{2-})$ / cm^{-1}
3	1062.4
4	1062.2
6	1061.4
7	1061.0
8	1060.2
Crystalline powders	1060.5

Table 2 Single particle measurement during a drying experiment. The numbers in the square brackets indicate the 95 % credible interval.

RH/%	m_0(400nm)	m_1	$\nu_1(CO_3^{2-})$ / cm^{-1}
67.6	1.425 [1.417, 1.437]	0.0236 [0.0196, 0.0276]	1061.31
62.8	1.430 [1.422, 1.438]	0.0228 [0.0196, 0.0260]	1061.20
56.6	1.438 [1.421, 1.446]	0.0248 [0.0208, 0.0296]	1060.84
49.2	1.441 [1.428, 1.454]	0.0252 [0.0208, 0.0288]	1060.51
43.3	1.440 [1.431, 1.454]	0.0260 [0.0220, 0.0292]	1059.99
38.2	1.442 [1.433, 1.451]	0.0272 [0.0240, 0.0308]	1059.59
33.1	1.446 [1.437, 1.460]	0.0268 [0.0228, 0.0308]	1059.11
29.6	1.451 [1.432, 1.461]	0.0260 [0.0212, 0.0312]	1058.55
27.6	1.446 [1.432, 1.460]	0.0268 [0.0220, 0.0312]	1058.26
25.4	1.453 [1.439, 1.468]	0.0272 [0.0216, 0.0328]	1057.68
23.2	1.457 [1.442, 1.472]	0.0284 [0.0232, 0.0340]	1057.39
20.8	1.462 [1.447, 1.477]	0.0264 [0.0208, 0.0324]	1056.93
19.1	1.456 [1.441, 1.472]	0.0288 [0.0232, 0.0336]	1056.37
17.5	1.461 [1.445, 1.476]	0.0276 [0.0220, 0.0344]	1056.00
16.1	1.461 [1.445, 1.477]	0.0272 [0.0216, 0.0324]	1055.88
15.1	1.466 [1.445, 1.476]	0.0280 [0.0228, 0.0340]	1055.94
14.1	1.470 [1.454, 1.480]	0.0280 [0.0228, 0.0340]	1055.64
13.3	1.470 [1.459, 1.486]	0.0280 [0.0228, 0.0332]	1055.23
12.6	1.473 [1.462, 1.484]	0.0272 [0.0228, 0.0328]	1055.18

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2 Initial water uptake during deliquescence

Broad band light scattering (BLS) experiments measure the intensity of light scattered by a single particle as a function of wavelength, λ. Resonances give rise to a ripple structure in the BLS spectrum (see Fig.1 in the main text). The position and the intensity profile of a specific resonance mode depend on the particle shape, the particle radius R (volume equivalent diameter for non-spherical particles), the wavelength of the light λ, and the refractive index $m(\lambda)$. Since the ratio of R and λ determines the resonance behaviour of the light within the particle it is convenient to define the size parameter $x = 2\pi N\lambda/R$ where N is the refractive index of the surrounding medium (here N=1).[1] The shift of the resonance position $\Delta\lambda$ indicates a change of R because the value of x at resonance remains constant. The change in R can then be calculated using Equation (1) under the assumption that the particle's shape and $m(\lambda)$ do not change appreciably. In the equation λ_R is the resonance position before the deliquescence starts.

$$\Delta R \sim \frac{R\Delta\lambda}{\lambda_R} \tag{1}$$

During the deliquescence experiment shown in Fig.8 in the main text (9.5 % $\leq$ RH $\leq$ 30 %) the peak positions in the BLS spectra drifts by $\Delta\lambda/\lambda_R = 0.5\%$. The particle size prior the efflorescence (at $t_E - 10$ ms) is R= 2070.5 [2052 2086] nm. Hence ΔR is $\sim$10nm.

References

1 C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles*, Wiley, Weinheim, Germany, Wiley Professional Paperback edn., 1983.