Supporting Information

Evaporative self-assembly of soft colloidal monolayers: The role of particle softness on microstructure

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Microgel particle characterization

The hydrodynamic diameter of the synthesised microgels with different crosslinking densities were determined using the dynamic light scattering (Nanopartica SZ-100, Horiba Scientific) technique. An exponential decay function is used to fit the autocorrelation function, $g^1(\tau)$, obtained from the DLS measurements. The fitting function used is: $y = A\ \exp(-x\Gamma) + y_0$.

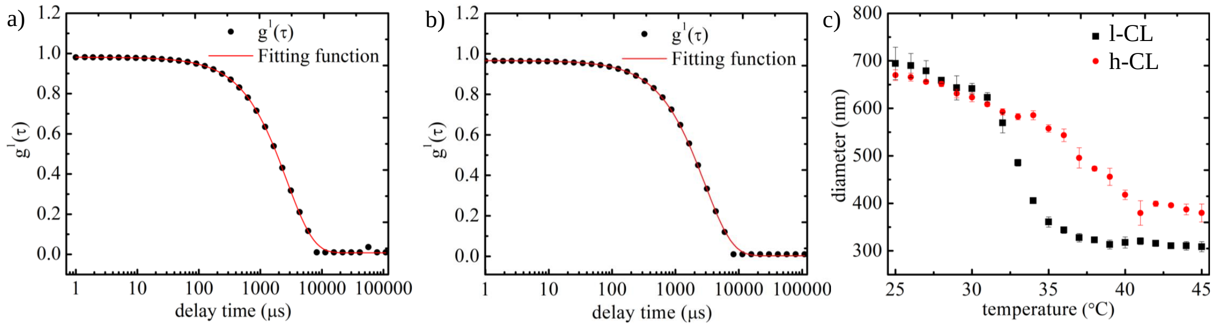

Figure S1: Electric field autocorrelation function vs. delay time for a dilute aqueous dispersion pf a) h-CL and b) l-CL microgel particles at 25°C. Red curves denote the exponential fit to the autocorrelation function. c) Particle diameters measured as a function of temperature for the l-CL (black squares) and the h-CL (red spheres) microgels.

In dispersions containing colloidal particles, the autocorrelation and delay time are related as [1], $g^1(\tau) = A\exp(-Dq^2\tau)+B$, where, $q = \frac{4\pi n}{\lambda}\sin(\frac{\theta}{2})$ is the scattering wave vector, n is the refractive index of the dispersion medium (n = 1.33), λ = 532 nm is the wave length of the laser, $\theta = 90°$ is the scattering angle and D is the diffusion coefficient. The best fit to the autocorrelation function gives Γ which can be used to compute the diffusion coefficient using, $D = \Gamma/q^2$. Further, the Stokes - Einstein relation is employed to obtain the hydrodynamic diameter of the particle. The DLS measurements were performed at temperatures ranging from 25 to 45°C, with the sample allowed to equilibrate for about 15-20 minutes at each temperature. The hydrodynamic diameter at each temperature is estimated from the average of five similar measurements.

Areal disorder

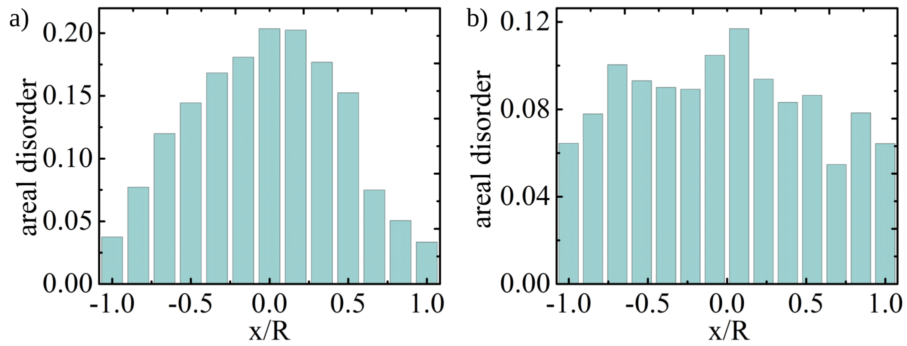

Figure S2: Evolution of areal disorder across the monolayer deposit of a) h-CL and b) l-CL microgels at their respective monolayer concentrations. The areal disorder is plotted as a function of x/R where 'R' is the radius of the drop and 'x' is the distance from the center of the drop.

References

[1] Puthusserickal A Hassan, Suman Rana, and Gunjan Verma. Making sense of brownian motion: colloid characterization by dynamic light scattering. *Langmuir*, 31(1):3–12, 2015.