

polymers in a reservoir at the same chemical potential as the colloid-polymer mixture. This corresponds to a contact potential of $7.0 k_B T$ per g/l of polymer reservoir concentration.

Determining the electrostatic interactions in non-aqueous colloidal dispersions is a non-trivial task [23, 34], as the charge and ionic strength are very small, although they still lead to significant interactions. The interactions have nevertheless been found to be well described by the screened Coulomb or Yukawa interaction, which reads

$$\beta u_{YUK}(r) = \begin{cases} \infty & \text{for } r < \sigma \\ \beta \epsilon \frac{\exp(-\kappa(r-\sigma))}{r/\sigma} & \text{for } r \geq \sigma \end{cases} \quad (2)$$

where r is the center to center separation of the two colloids. The contact potential is given by

$$\beta \epsilon = \frac{Z^2}{(1 + \kappa\sigma/2)^2} \frac{l_B}{\sigma}, \quad (3)$$

where Z is the colloid charge, κ is the inverse Debye screening length and l_B is the Bjerrum length.

One may thus assume interactions between the colloids take the form of a short-range ‘depletion’ attraction and long-ranged screened Coulomb repulsion, and a nearly hard core. Both have been measured in nearly identical systems and found to be in good agreement with theory [23, 24]. Here we have determined the colloid charge and ionic strength by comparison of the measured $g(r)$ with one obtained from Monte Carlo simulation according to a Yukawa potential [24] in the ergodic fluid part of the state diagram ($\phi = 4.8 \times 10^{-4} \pm 2 \times 10^{-5}$, $c_p = 0$) to be $Z \approx 600 \pm 200 e$ where e is the elementary unit of charge, and the inverse Debye length $\kappa\sigma \approx 1 \pm 0.4$. The radial distribution function $g(r)$ fitting is shown in SFig. 1. This method is found to be consistent with electrophoretic measurements [24]. The values we obtain for the colloid charge are somewhat (up to 25%) higher than those quoted previously in literature where samples were prepared using dry colloids [24, 34]. For these parameters mode-coupling theory predicts vitrification at higher volume fractions [25]. However, this analysis poses a significant question: the contact potential between two such colloids is $\beta \epsilon \sim 1000$. This is much greater than the strength of the attraction induced by the polymer. Throughout the measurements we find that clustering of gelation occurs at a polymer concentration around $c_p = 4.0 \text{ gl}^{-1}$, which corresponds to a contact potential of around $30 k_B T$ [Eq. (1)].

We are reasonably confident about our understanding of the attraction induced by the polymer [23]. We speculate that the charge is in fact different between the isolated colloids