Supplementary information for article: Extreme mechanics of colloidal polymers under compression: buckling, creep and break-up

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I. OPTICAL TWEEZER DETAILS

A. In-plane rotational freedom

The focused laser spots form harmonic traps at the image focal plane [1]. We checked that the trapped particles retained in-plane rotational freedom by using fluorescence microscopy in which only the dyed patches remain visible, Fig. 1. Indeed, in both traps, the position of the patches and the associated particle orientation, ϕ, rotate over time, Fig. 1(b).

To quantify the rotational diffusion, we tracked the positions of the patches over time using tracking software and calculated the time-dependent particle orientation $\phi(t)$. From this, we determined the mean-square rotation, $\langle\Delta\phi^2\rangle$, as shown in Fig. 1(c). For both traps the data fits well with a diffusive power law $\langle\Delta\phi^2\rangle = 2D_r\Delta t$ with rotational diffusion constant D_r, confirming the free rotation of the particle. From the best fit we obtain $D_r = 0.05(5)$ rad^2/s, a factor of two higher than the expected Stokes-Einstein diffusion coefficient $kT/8\pi r^3\eta = 0.02$, where the viscosity $\eta = 1.9$mPas for the binary mixture [2]. This deviation could originate from hydrodynamic effects due to the vicinity of the capillary wall, the assumption of no-slip boundary conditions or the composite nature of the particle [3].

B. Optical tweezer callibration

We calibrated the traps by tracking the Brownian movement of a single colloidal dipatch particle in the trap. Such calibrations were performed after each experiment to account for slight differences in laser intensity between measurements. Because of the weak laser intensity ($\sim$ 5 mW) and resulting soft trap, conventional back focal plane interferometry was not a convenient trap callibration technique. Instead, the trapped particle was imaged directly in real space using bright-field microcopy, and the center of the trapped particle was localized in the imaging plane in the same way using particle tracking software as particles of the chain. The overdamped dynamics of a trapped colloidal particle at position (x, y) from the trap center is described by [1]:

$$\dot{x} = -\frac{k}{\gamma}x + \sqrt{\frac{2kT}{\gamma}}\xi(t), \tag{1}$$

with k the spring constant of the trap, γ the drag coefficient and $\xi(t)$ a normalized Gaussian stochastic process. This equation is solved by the mean-square displacement (MSD)

$$\langle \Delta x^2 \rangle = 2\left(1 - e^{-\Delta t/\tau}\right)\frac{kT}{k}, \tag{2}$$

yielding an exponential relaxation with time constant $\tau = \gamma/k$ where Δt is the lag time. Equivalently, Eq.1 can be solved in terms of the power spectral density (PSD) which follows the Lorentzian [4]

$$P(f) = \frac{kT}{\gamma\pi^2 f^2[1 + (f_c/f)^2]}, \tag{3}$$

where f is frequency and $f_c = 1/2\pi\tau$ the roll-off frequency. Fig. 1(d,e) shows the MSD and PSD of a dipatch colloidal particle in the static optical trap and a fit with Eqs. 2 and Eqs. 3, respectively. The good quality of the fits proves that the tweezers are well described by harmonic traps. From the best fits of both the PSD and the MSD, we obtain a diffusion coefficient $D = 0.03 \pm 0.001$ μm^2/s and a trap spring constant $k_s = 0.6 \pm 0.01$ pN/μm. This diffusion constant is consistent with the diffusion constant for freely diffusing particles, $D = 0.035 \pm 0.05$ μm^2/s that was measured by tracking an ensemble of free particles. The spring constant also agrees well with simply fitting a gaussian to the histogram of displacements to obtain $\sigma = 0.08$ μm, and $k_s = kT/\sigma^2 = 0.58 \pm 0.02$ pN/μm, see Fig. 1(f). Similar calibrations were done after each experiment for the static trap with slight variations in the found trap constants on the order of 0.1 pN/μm.

While the static trap was always used to infer the force exerted on the chain, a calibration of the mobile trap was also performed. Unfortunately, during experiments no measurements of stationary single particles in the mobile trap were recorded; therefore an indirect method was used, which is based on the calibration of the stationary trap. Force-versus-time curves of the mobile trap calculated for varying spring constants were fitted against the known force-time curve of the stationary trap, see Fig 2. The optimal spring constant was chosen as the one yielding the smallest deviation (smallest sum of squared differences, $\sum(F_s - F_m)^2$) between the two force curves. For the best fit, we obtain $k_m = 0.42 \pm 0.05$ pN/μm, where the uncertainty was estimated by visual inspection. The method assumes that, on average, there is a force balance between the two ends of the chain, such that the stationary and mobile trap, on average, exert equal but opposite forces. This assumption should hold under quasi-static conditions with a low trap speed. This mobile-trap spring constant, together with the spring constant of the static trap, is used to determine the effective spring constant k_{eff} of

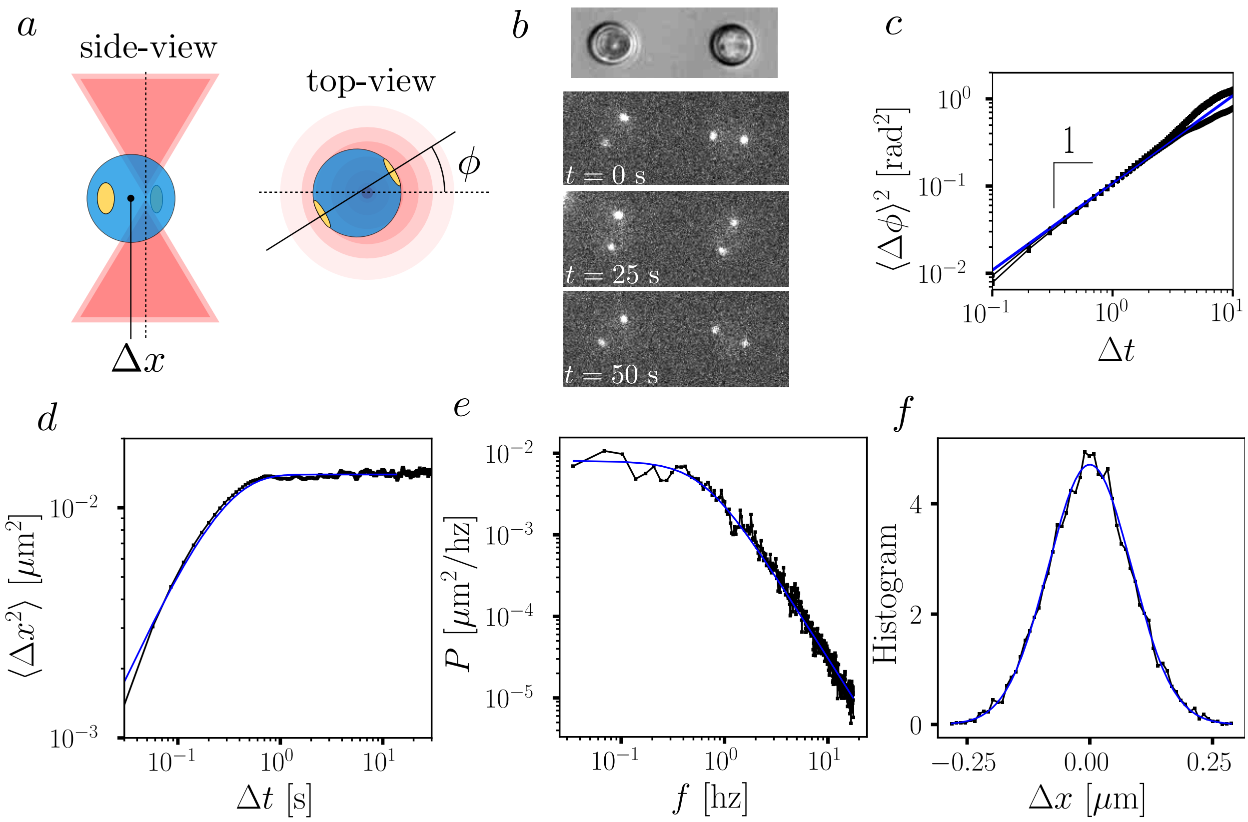

FIG. 1. Trap calibration of the static optical tweezer (a) Schematic of a dipatch particle in an optical trap (b) Top: Bright field image of trapped particles. Bottom: Three fluorescence images showing the dyed patches diffusing over time (c) Mean-square rotational displacement and diffusive power law fit $\langle \Delta\phi^2 \rangle = 2D_r\Delta t$. (d) Mean-square displacement of the center of a trapped particle and best-fit exponential relaxation, imaging was done at 35 fps for 5 min (e) Power spectral density and best-fit Lorentzian. (f) Histogram of particle positions in the x-direction and Gaussian fit.

the two traps in series as described in the manuscript. To infer the force exerted on the chain, however, the static trap is always used as this can be done more reliably.

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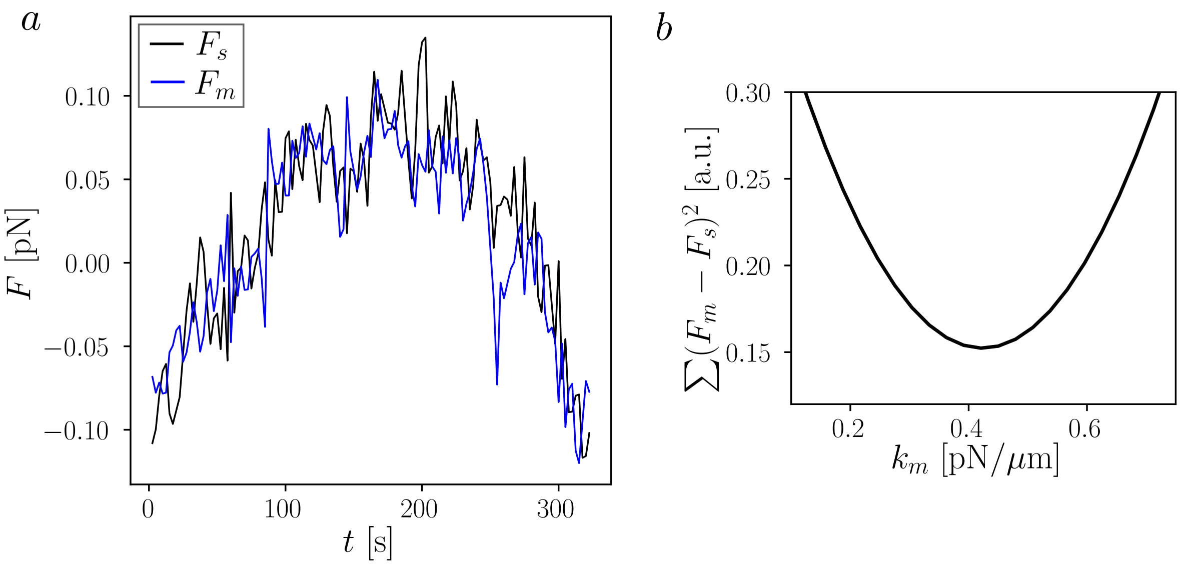

FIG. 2. Trap calibration of the mobile optical tweezer (a) Force exerted by the static optical (F_s) and mobile optical trap (F_m) during a single experiment. Here $k_m = 0.42$ pN/μm was used. (b) The sum of squares of the difference between the force of the mobile and the static optical tweezers. A least-squares fit gives $k_m = 0.42$ pN/μm.

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