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Supplementary Materials for

Guiding kinetic trajectories between jammed and unjammed states in 2D colloidal nanocrystal-polymer assemblies with zwitterionic ligands

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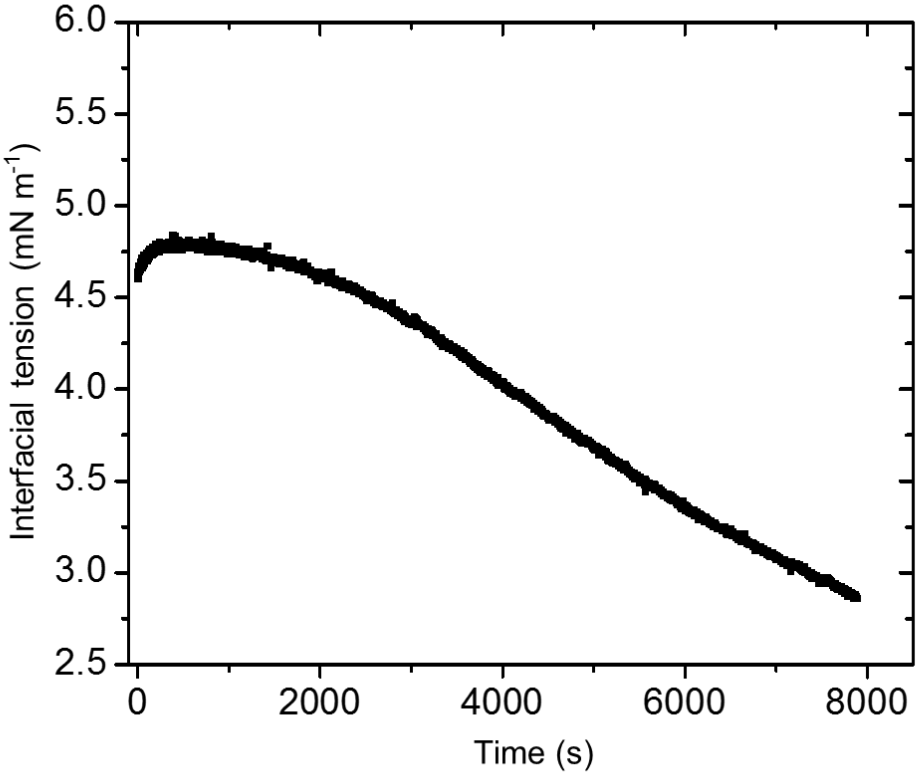

Fig. S1. Interfacial tension as a function of time for a system configured with both Fe_3O_4 NCs and 3K-PDMS-NH_2. A pendant droplet of DMF with 0.5 mg mL^{-1} Fe_3O_4 NCs is suspended in a bath of PDMS/dodecane with 5% *w/w* 3K-PDMS-NH_2. After NCPS monolayer formation reached steady state, the interfacial tension decreased to $\gamma_{polar/non\text{-}polar}$ = 2.85 mN m^{-1}.

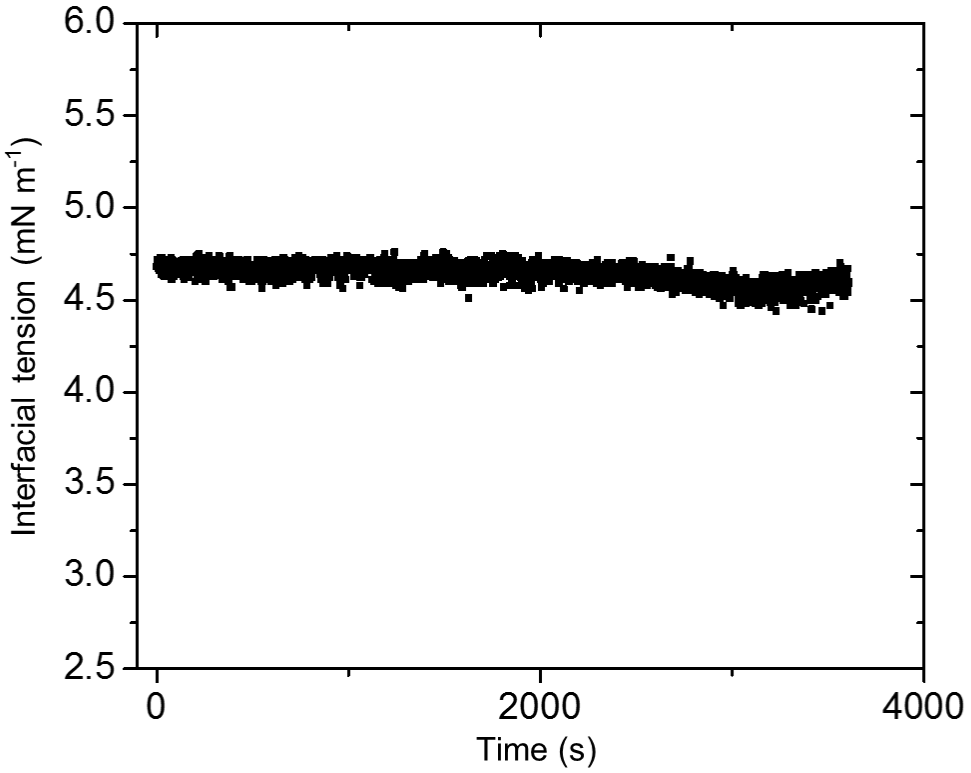

Fig. S2. Interfacial tension as a function of time for a system without NCPSs. A pendant droplet of DMF suspended in a bath of PDMS/dodecane. Without introducing anything in both phase, the system keeps at a steady state, the interfacial tension is 4.65 mN m^{-1}.

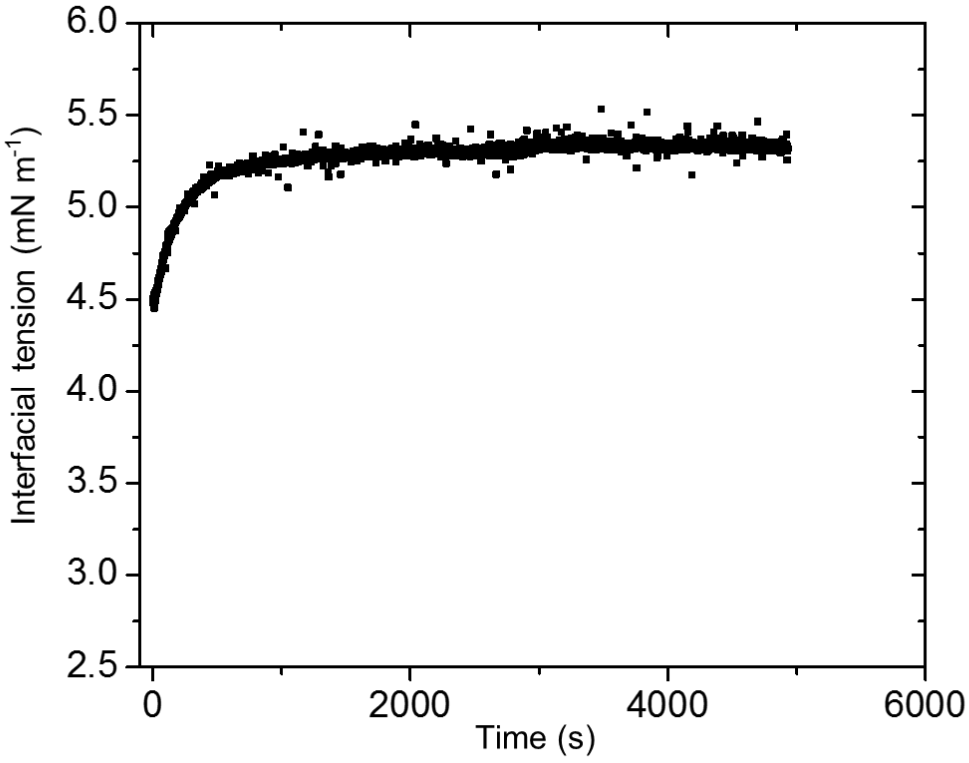

Fig. S3. Interfacial tension as a function of time for a system configured with only 3K-PDMS-NH_2. A pendant droplet of pure DMF suspended in a bath of PDMS/dodecane with 5% *w/w* 3K-PDMS-NH_2. After the system reached steady state, the interfacial tension increased to $\gamma_{polar/nonpolar} = 5.25$ mN m^{-1}.

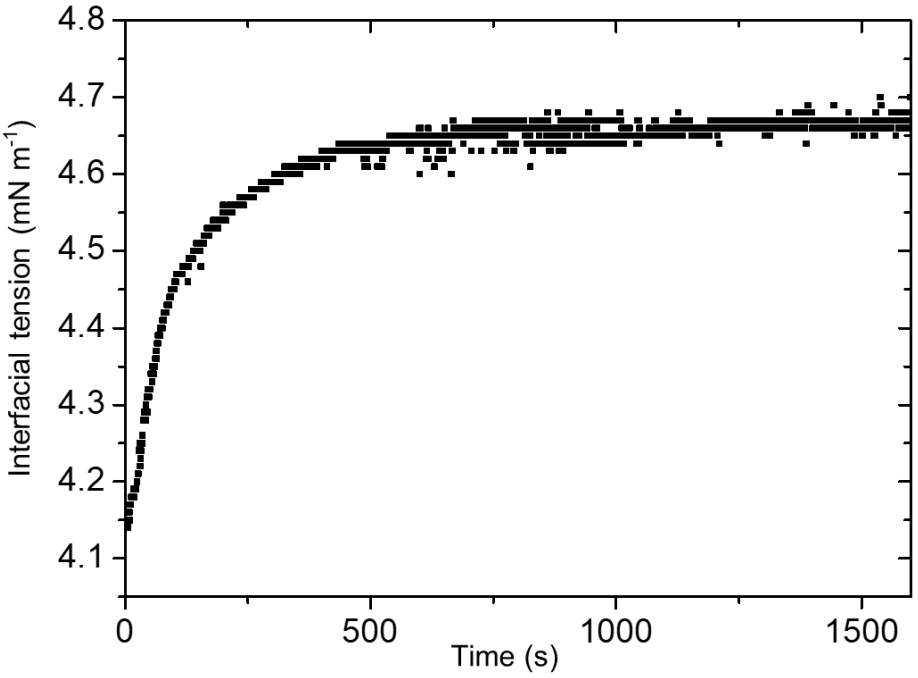

Fig. S4. Interfacial tension as a function of time for a system configured with only zwitterionic ligand 1. A pendant droplet of DMF with 0.5 mg mL^{-1} Ligand **1** suspended in a bath of PDMS/dodecane. After the system reached steady state, the interfacial tension increased to $\gamma_{polar/nonpolar} = 4.67$ mN m^{-1}.

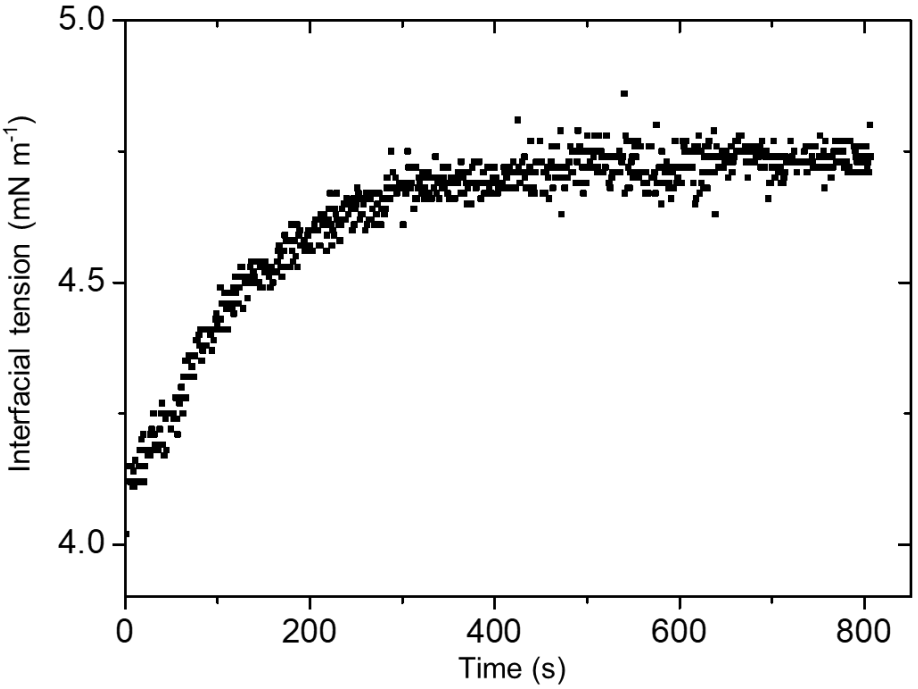

Fig. S5. Interfacial tension as a function of time for a system configured with zwitterionic ligand 1 and 3K-PDMS-NH_2. A pendant droplet of DMF with 0.5 mg mL^{-1} Ligand **1** suspended in a bath of PDMS/dodecane with 5% *w/w* 3K-PDMS-NH_2. After the system reached steady state, the interfacial tension increased to $\gamma_{polar/nonpolar}$ = 4.74 mN m^{-1}.

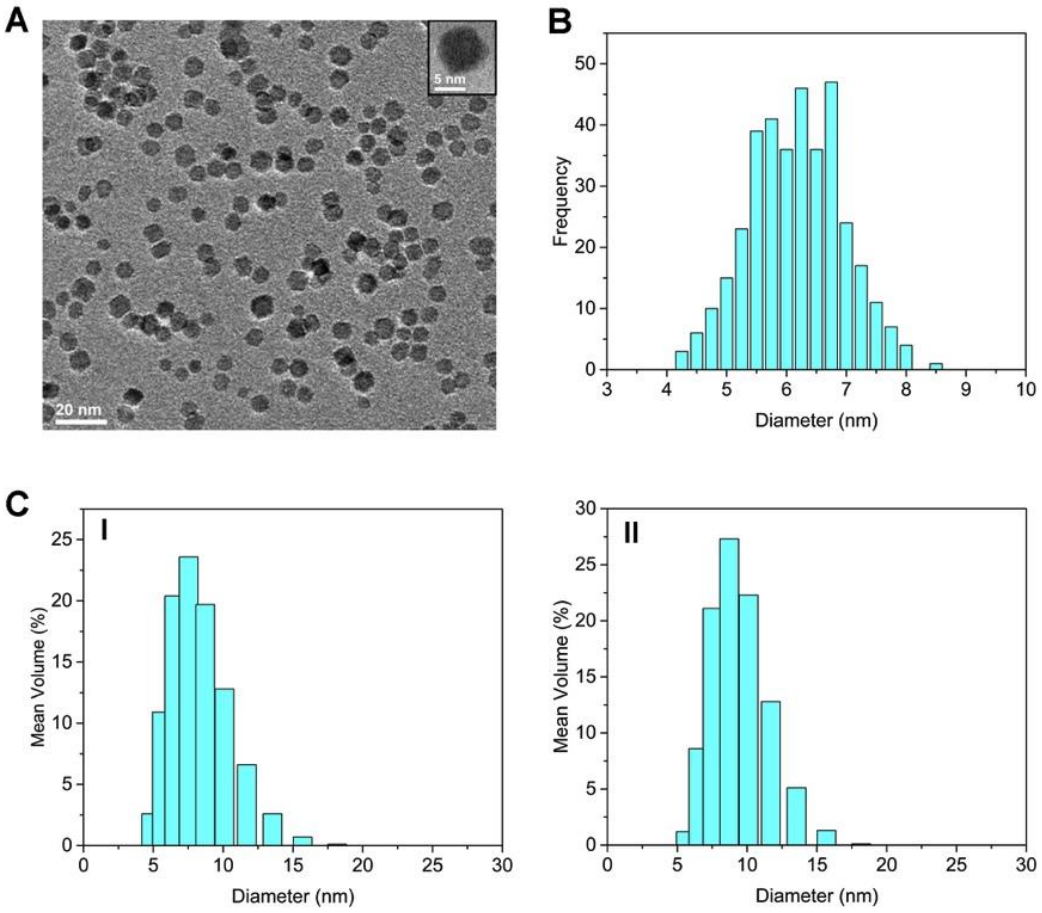

Fig. S6. Zwitterionic ligand binding to naked NC surfaces. (**A**) TEM of naked Fe_3O_4 NCs; **(B**) Size-Distribution of naked Fe_3O_4 NCs calculated from TEM images (sample size ~400 NCs); (**C**) Size-Distribution of naked Fe_3O_4 NCs dispersed in DMF calculated from DLS data in the absence (**I**) or in the presence of zwitterionic ligand **1** (**II**).

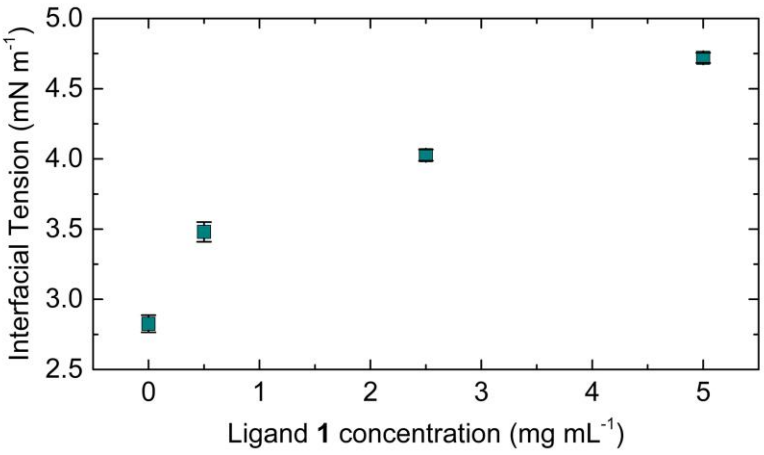

Fig. S7. Steady-state interfacial tension as a function of zwitterionic ligand concentration. Interfacial tension at the onset of wrinkling in interfacial films for pendent droplets of DMF suspended in a bath of PDMS/dodecane for systems configured with 3K-PDMS-NH_2 (5% *w/w*), Fe_3O_4 NCs (0.5 mg mL^{-1}), and varying concentrations of zwitterionic ligand **1** (0–5.0 mg mL^{-1}). Small standard deviations indicate excellent reproducibility.