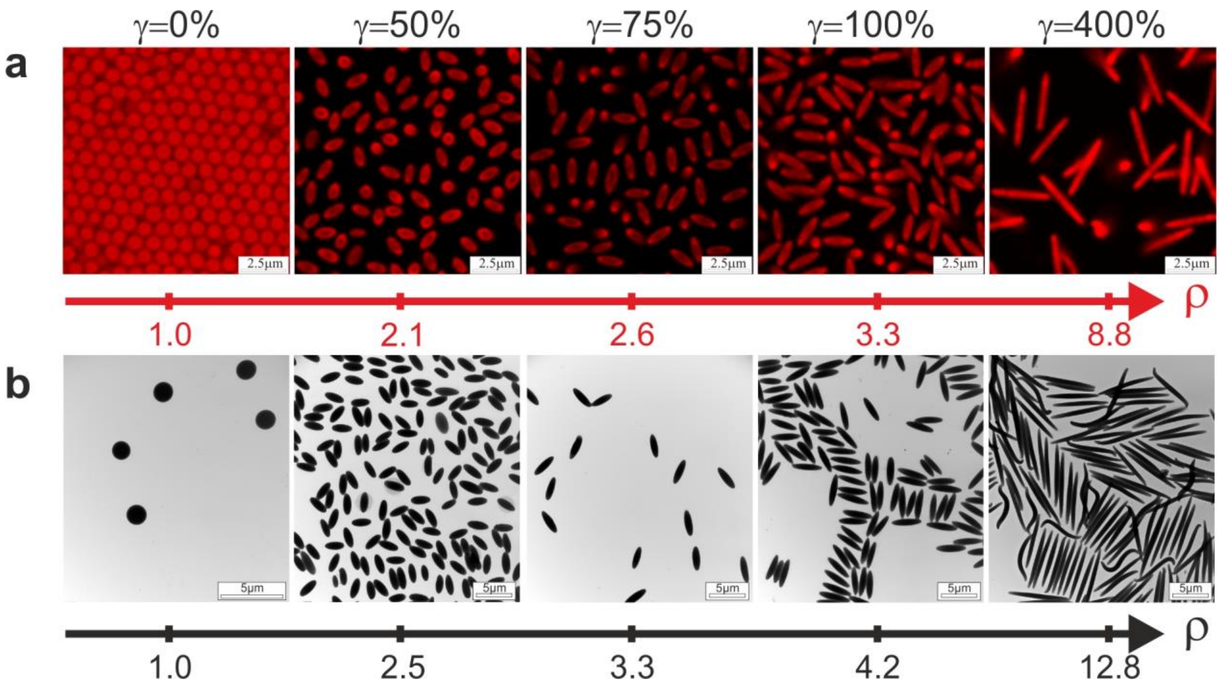
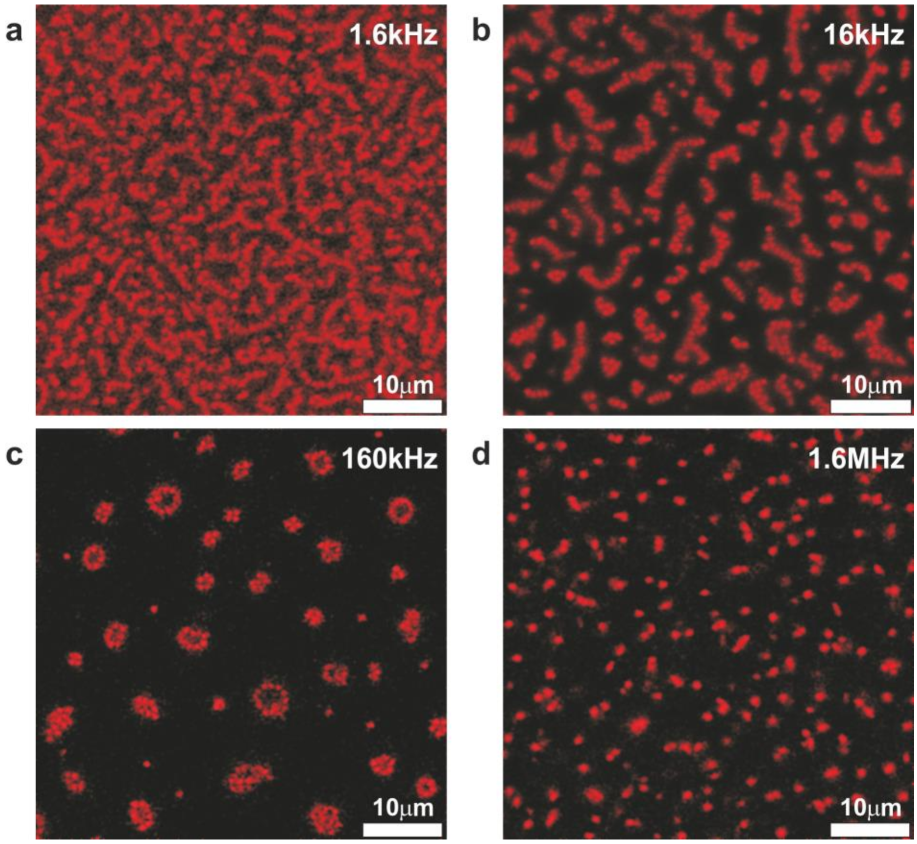
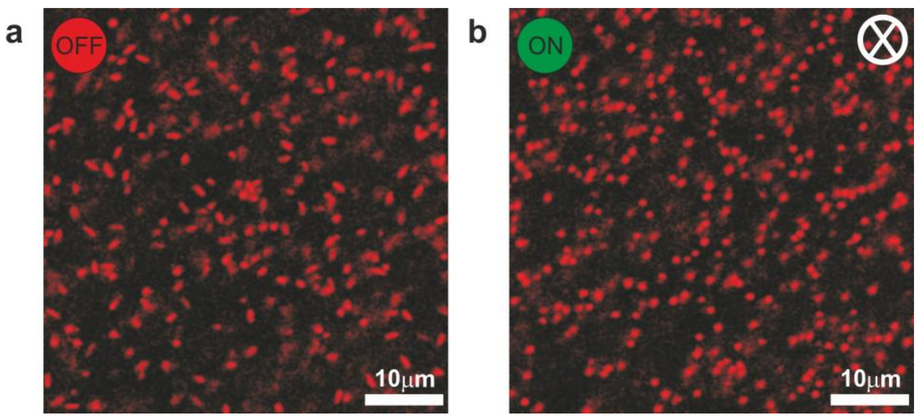
# Shape matters for a new colloidal self-assembly mechanism for synthetic microtubules




**Supplementary Fig. 1 | Microscopy of composite ellipsoidal microgels synthesized with different aspect ratios.** Microscopic characterization of the composite microgel morphologies obtained at different uniaxial deformation ($\gamma$). CLSM (**a**) and TEM micrographs (**b**) of the core-shell particles at the water/glass interface and in air on carbon-coated copper grids, respectively. The evolution of the aspect ratio ($\rho$) determined from the analysis of the different micrographs with the two different techniques is indicated as a function of the deformation.




**Supplementary Fig. 2 | Frequency dependence of the self-assembly.** CLSM *xy*-images showing the assemblies formed by a 1 wt% ellipsoidal composite microgel dispersion ($\rho$ = 3.3) in response to an applied AC electric field $E$ = 125 kV $m^{-1}$ as a function of frequency. The applied range of frequencies varied from $f$ = 1.6 kHz to 1.6 MHz. Representative images show the formation of (**a**) solid worm-like lattices formed in the vicinity of the electrodes at $f$ =1.6 kHz, (**b**) "dynamic" worm-like lattices which extend over 10 µm in the z-plane at $f$ = 16 kHz, (**c**) tubular structures at $f$ = 160 kHz, and (**d**) a partially aligned fluid at $f$ = 1.6 MHz. All structures formed are fully reversible, and the colloidal dispersions relax to their initial fluid-like disordered state after the field is turned off.




**Supplementary Fig. 3 | Salt effect on the dipolar association**. The experiment was performed on a 1 wt% ellipsoidal dispersion ($\rho$ = 3.3) containing 1 mM KCl under an AC electric field $E$ = 125 kV $m^{-1}$ with a frequency, $f$ = 160 kHz. The CLSM *xy*-images correspond to (**a**) randomly orientated ellipsoids (disordered particle fluid), (**b**) oriented ellipsoids with their major axes aligned in the field direction (aligned particle fluid).

**Supplementary Table 1**

| $\gamma$ | 0% | 50% | 75% | 100% | 400% |
|---|---|---|---|---|---|
| $2a$ [nm] CLSM | 824 ± 47 | 1276 ± 81 | 1528 ± 88 | 1770 ± 117 | 3308 ± 176 |
| $2b$ [nm] CLSM | 824 ± 47 | 614 ± 50 | 591 ± 38 | 542 ± 42 | 381 ± 26 |
| $\rho$ (CLSM) | 1 | 2.1 ± 0.1 | 2.6 ± 0.2 | 3.3 ± 0.2 | 8.8 ± 0.7 |
| $2a$ [nm] TEM | 623 ± 31 | 1149 ± 58 | 1401 ± 71 | 1634 ± 90 | 3408 ± 267 |
| $2b$ [nm] TEM | 623 ± 31 | 459 ± 28 | 424 ± 22 | 392 ± 28 | 270 ± 32 |
| $\rho$ (TEM) | 1 | 2.5 ± 0.1 | 3.3 ± 0.2 | 4.2 ± 0.3 | 12.8 ± 1.6 |
| $2a_c$ [nm] TEM | 534 | 986 | 1184 | 1388 | 2920 |
| $2b_c$ [nm] TEM | 534 | 393 | 359 | 331 | 228 |
| $\rho$ (TEM) | 1 | 2.5 | 3.3 | 4.2 | 12.8 |
| $t_a$ [nm] | 145 | 145 | 172 | 191 | 194 |
| $t_b$ [nm] | 145 | 111 | 116 | 105 | 76 |
| $R_h$ at 20°C [nm] (DLS) | 537 | 499 | 501 | 527 | 670 |
| $R_h$ at 50°C [nm] (DLS) | 348 | 333 | - | 428 | 493 |
| $Q_{eff}$ at 20°C [$e^-$] | 2104 | 1779 | 1601 | 1767 | 2148 |

**Supplementary Table 1 | Summary of the main characterization results.** Average dimensions and aspect ratios of the composite microgel particles stretched at a deformation of $\gamma$ = 0%, 50%, 75%, 100% and 400% determined from statistical analysis of CLSM and TEM micrographs. The notations $2a$, $2b$ refer to the long and short full axis of the particles and $2a_c$, $2b_c$ to the long and short full axis of the PS core. $t_a$ and $t_b$ indicate the PNIPMAm shell thickness along the long and short axis, and $\rho$ the determined aspect ratio. This table further includes the apparent hydrodynamic radius $R_h$ measured by DLS at 20°C and 50°C and the effective charge of the different particles $Q_{eff}$ estimated at 20°C from the combination of DLS and electrophoretic measurements of the different core-shell particles.