Advanced Materials

Directing colloidal self-assembly with biaxial electric fields

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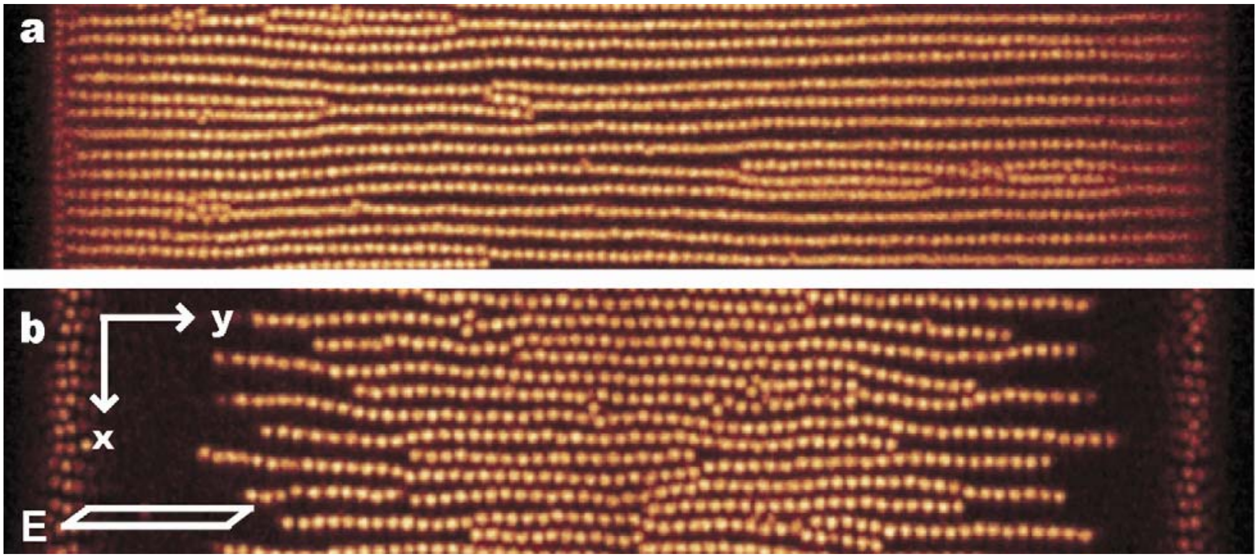
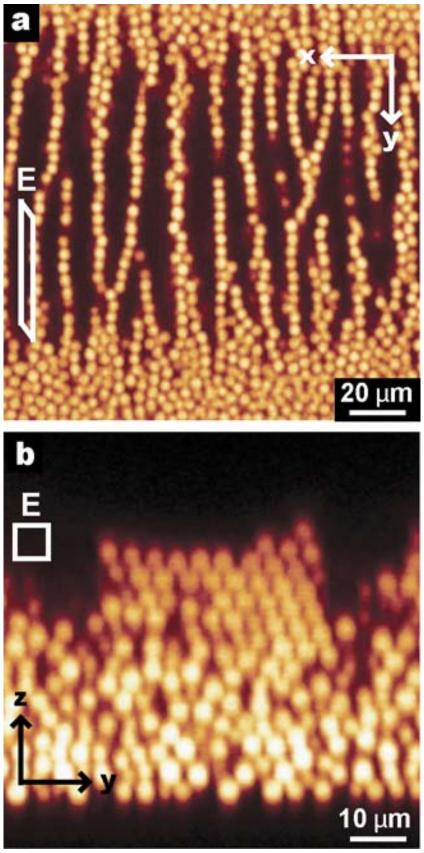
Supplementary Figure S1



Overview of the entire width of the sample space (~ 200 µm) in order to identify dielectrophoretic effects. The sample space was filled with a suspension of 2.00 µm diameter PMMA-particles in a density-matched CHB-decalin mixture, with particle volume fraction $\varphi = 0.20$ and $\beta = -0.22$ ($\varepsilon_p < \varepsilon_m$). The confocal xy images were taken (a) in the center of the sample space, at $z = 0$, and (b) approximately 10 µm above the bottom of the sample space ($z = -90$ µm). The orientation of the plane of the biaxial electric field 'E', here perpendicular to the imaging plane, is indicated with a skewed white rectangle ($\Delta V = 230$ V). The fact that in (b) the particles formed a dense band in the middle of the cell, while in (a) the sheets extended over the entire width, indicates that (only) near the corners of the cell the particles experienced an inward dielectrophoretic force. We found that the sheets spanned the entire cell width starting at 60 µm from the bottom and top wall. This agrees well with the calculated electric field profile.

Supplementary Figure S2



Suspension with a positive dielectric constant contrast ($\varepsilon_p > \varepsilon_m$) in a biaxial electric field ($\Delta V = 260$ V). a, *xy* image ($z = -80$ µm). b, *yz* image (halfway the length of the sample cell). The sample space was filled with a suspension of PMMA particles in pure *cis*-decalin, $\varphi = 0.20$. Because the polarizability in this solvent was rather small, $\beta = +0.06$, we used 3.9 µm diameter particles to obtain sufficiently strong dipole interactions. These large particles did not display much Brownian motion, though, and formed a dense sediment of ~60 µm thick on the bottom of the sample cell due to the large density mismatch (PMMA ~ 1.19 gml^{-1}, decalin ~ 0.89 gml^{-1}). This hindered the restructuring in the biaxial electric field, therefore giving rise to less well ordered structures than we previously observed for our density-matched negative contrast suspensions ($\varepsilon_p < \varepsilon_m$) of 2.00 µm diameter particles. Nevertheless, the particles were still seen to form the familiar field-aligned hexagonal sheets, spanning the middle of the cell. As can be seen in panel (a), many particles collected at the side walls when the electric field was applied; dielectrophoretic forces pushed the particles outward due to the inverted dielectric constant contrast as compared to the case in Supplementary Figure 1. Unfortunately, the sedimentation of the particles prevented us from using the center of the sample space, where the electric field is homogeneous. Slow rotation of the sample cell in the gravitational field may already solve this problem, though.

Supplementary Movies

Movie S1. XY recording of the structural evolution of a suspension in a biaxial field oriented along the yz plane. Shown are the initial stages of the structural evolution of the suspension in the center of the sample space after switching on ΔV = 230 V at t = 0. The frames are 55 x 55 μm^2 and the movie covers 5 minutes and 40 seconds.

Movie S2. YZ recording of the structural evolution of a suspension in a biaxial field oriented along the yz plane. Shown are the initial stages of the structural evolution of the suspension after switching on ΔV = 230 V at t = 0. Note that the particles have an elongated appearance due to the asymmetric point-spread function in the z direction. The frames are 55 x 55 μm^2 and the movie covers 3 minutes.

Movie S3. Switching between uniaxial and biaxial fields. *xy* recording of the evolution of the suspension structure in the center of the sample space during the switching sequence 'zero field – uniaxial field – biaxial field – uniaxial field – zero field' (ΔV = 230 V). The field was switched at t = 10 seconds (zero field → uniaxial), t = 250 seconds (uniaxial → biaxial), t = 335 seconds (biaxial → uniaxial) and t = 420 seconds (uniaxial → zero field). The uniaxial field was oriented along the z direction and the biaxial field along the *yz* plane (i.e., both were perpendicular to the imaging plane). The frames are 55 x 55 μm^2.

Movie S4. Switching between uniaxial and biaxial fields. Same switching sequence as in Movie S3, but now as a yz recording in which both the uniaxial and biaxial fields are parallel to the imaging plane. Note that the particles have an elongated appearance and can not always be resolved individually due to the asymmetric point-spread function in the z direction.The frames are 55 x 55 μm^2.

Movie S5. Self-supported permanent sheet structures– *xy* recording. Sheets of particles, which formed at ΔV = 260 V, were thermally annealed. The confocal microscopy movie shows how, after switching off the electric field, the structures remained intact and free-standing. There is no motion of the individual particles visible (compare with Supplementary Video 1), except for a number of freely suspended particles that were not incorporated into the sheets. The change in intensity during the recording is due to bleaching of the fluorescent dye in the particles. The frames are 160 x 160 μm^2 and the movie covers 90 seconds.