

*Supporting Information*

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# Multidirectional Colloidal Assembly in Concurrent Electric and Magnetic Fields

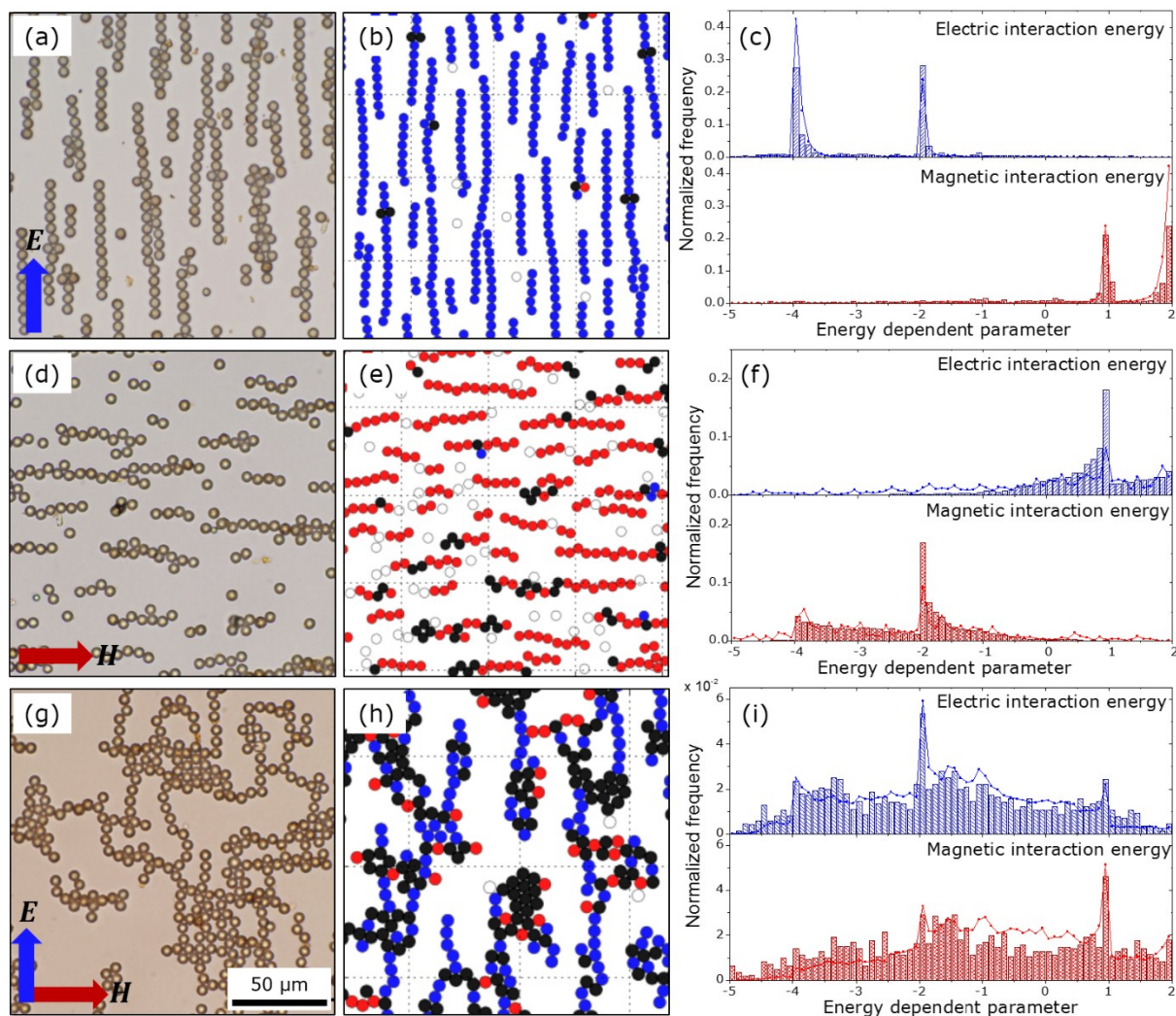
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**Figure S1.** Comparison of structures formed in experiments and simulations. (a, d, g) Experimental microscope images for microparticle bidirectional chains formed after 12 s of field application at (a)  $E = 20$  V/mm,  $H = 0$  A/m; (d)  $E = 0$  V/mm,  $H = 90$  A/m; and (g)  $E = 20$  V/mm,  $H = 100$  A/m. (b, e, h) Brownian dynamics simulation snapshots of particle structures assembled at (b)  $\mu^{\text{el}} = 0.3$ ,  $\mu^{\text{mag}} = 0$ ; (e)  $\mu^{\text{el}} = 0$ ,  $\mu^{\text{mag}} = 0.35$ ; (h)  $\mu^{\text{el}} = 0.45$ ,  $\mu^{\text{mag}} = 0.45$ . (c, f, i) The quantitative comparison of the particle structures observed in experiments (bars) and simulations (lines).

In order to investigate the validity of our simulations, we present here three extreme cases of the microparticle assembly process: (1) Only electric field,  $E = 20$  V/mm,  $H = 0$  A/m; (2) Only magnetic field,  $E = 0$  V/mm,  $H = 90$  A/m; and (3) High intensity electric and magnetic field,  $E = 20$  V/mm,  $H = 90$  A/m. The particle structures formed under these three assembly conditions are shown in Figure S1a, d and g. The BD simulations for similar field conditions were performed and the snapshots are provided in Figure S1b, e and h. As can be observed, in the presence of only electric field fine particle chains are formed both in experiments and

simulations. The structures show two distinct peaks in the energy distributions, corresponding to the particles present within the vertical chains and at the chain terminals (Fig. S1c). However, the particle chains formed by magnetic field are partially staggered (Fig. S1d) due to dislocation of the center of dipole from the center of mass of the particle (for details see section 2.3). Here BD simulations performed with off-centered dipole ( $\delta = 0.2\sigma$ ) show similar particle assembly (Fig. S1e and f) and validate the applicability of our simulation procedure. The third case of strong electric and magnetic field, in both experiments and simulations shows the formation of a coarse particle network (Fig. S1g and h). Here the electric and magnetic energy parameters for experiments and the simulations show a broad distribution and lack of any distinct peaks (Fig. S1i). This observation can be attributed to the presence of coarse network formed by multiparticle diameter structures (partially collapsed) over single particle chains.