

Supporting Information for the article:

Magnetically reconfigurable colloidal patterns arranged from arrays of self-assembled microscopic dimers

Experimental Details.

Magnetic colloids and dipolar interactions. The magnetic colloids used are monodisperse polystyrene particles from Invitrogen (Dynabeads Myone) with carboxylic surface groups and doped with small superparamagnetic iron oxide grains (size from 10 Å to few nm). The particles have diameter $D = 1.05 \mu\text{m}$, density $\rho = 1.8 \text{ g/cm}^3$ and magnetic volume susceptibility $\chi \sim 1$. In the presence of an applied magnetic field \mathbf{H} , the particles become magnetized and acquire a dipole moment \mathbf{m} , pointing along the field direction, $\mathbf{m} = V\chi\mathbf{H}$ where V is the particle volume. The interaction between two equal dipoles separated by a distance r , scale as $U_{dip} \sim m^2(1 - 3\cos^2\alpha)/r^3$, being α the angle between the dipoles and their line of separation. Thus, when $\alpha < \alpha_{magic}$, $U_{dip} < 0$ and the dipoles attract each other, while when $\alpha > \alpha_{magic}$, $U_{dip} > 0$ and the dipoles repel each other, being $\alpha_{magic} = 54.7^\circ$, the "magic" angle when $U_{dip} = 0$. Note that in Fig. 3 of the article, the orientation angle of the dimers ϑ is related to α by $\alpha = \pi/2 - \vartheta$. The particles, dispersed in water, were electrostatically stabilized by the negative charges acquired from the dissociation of the surface carboxylic groups (COO⁻). The original aqueous suspension of the particles (10 mg/ml – $7 \cdot 10^9$ beads/ml) was diluted with high deionized water (18.2 MW·cm, MilliQ system) up to a density of $\sim 10^7$ beads/ml and few droplets were deposited on top of the FGF coated with the photoresist. Due to gravity, after a few minutes, all particles sediment above the film, and remained confined above the photoresist without sticking to it.

Coating of the Garnet Film. The positive Photoresist AZ-1512 (Microchem, Newton, MA) was applied on top of the FGF by using spin coating (Spinner Ws-650Sz, Laurell) and UV irradiation (Mask Aligner MJB4, SUSS Microtec). The procedure used to coat the FGF with a film thick 1.7 micron consists in the following steps. The FGF was clean for 15 min. in an ultrasonic bath filled first in acetone (Merck) and then in isopropanol (Sigma), and finally the film was dried under stream of N₂. Above the clean FGF, few drops of photoresist were deposited by using a pipette, and dispersed above the film in the spin coater at 3000 rpm for 30s. The FGF film with the photoresist was then placed on a hot plate at 95o for 1min (softbake) and then subjected to 5s of UV irradiation at a power of 30 mW/cm² in order to crosslink the photoresist. A final post-bake process was applied by placing the film above a hotplate at 115°C for 50 s.

Observation of the paramagnetic dimers and external field. The particle dynamics was recorded by using a 1.3 megapixel monochrome camera (Pixelink, PL-A741) working at 23.1 fps for a field of view of $68 \times 51 \mu\text{m}^2$. The camera was mounted on a light microscope (Nikon E400) equipped with a 100×, 1.43 numerical aperture CFI Plan Fluor oil immersion objective. The average distance/orientation of the particles were measured by using particle tracking. In particular, the positions of the particles in the plane were obtained from the analysis of .AVI videos recorded via a commercial software (STREAMPIX) and later on analyzed by using a home-written program based on MATLAB. The DC magnetic field applied to the garnet film was provided by using two custom-made Helmholtz coils arranged perpendicular to each other and having the

main axes along the (x,y) directions. The two coils were connected to a DC power supply (TTi El 302).

Supporting Videos.

Video 1 (.AVI): thermal motion of an ensemble of paramagnetic dimers assembled above a ferrite garnet film and in absence of an external magnetic field, Fig. 1 of the manuscript.

Video 2 (.mpeg1): parallel dimers arranged into an herringbone pattern and subjected to an oscillating magnetic field of strength $H = 500$ A/m, frequency $\nu = 1$ Hz and applied parallel to the BWs. During oscillations, the pattern reversibly changes configuration and the dimers along the same BW have the same average orientation, Fig. 2(b) of the manuscript.

Video 3 (.mpeg1): transition from a pattern characterized by parallel dimers to branched chains due to the application of an external magnetic field parallel to the BWs and of strength $H = 800$ A/m, Fig. 2(c) of the manuscript.

Video 4 (.mpeg1): transition from a phase characterized by parallel dimers to buckled chains due to the application of an external field perpendicular to the FGF, of strength $H = 1000$ A/m, Fig. 2(f) of the manuscript. The field is periodically switch on and off to show the reversibility of the pattern.

Video 5 (.mpeg1): Diffusive motion of polystyrene non-magnetic particles above a FGF film, Fig. 3(a) of the article.

Video 6 (.mpeg1): Diffusive motion of polystyrene non-magnetic particles above an ensemble of paramagnetic particles assembled into buckled chains by an external field of strength $H = 600$ A/m, Fig. 3(b) of the article.