Supporting Information

Magnetic Assembly of Polymer Colloids in Ferrofluid and Its Display Applications

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Figure S1. Experimental setup for measuring the reflection spectra of magnetically controlled display unit. The magnetic field with controlled strength was created by electromagnet and DC power source. Reflection spectra were measured by Ocean Optics spectrometer.



Figure S2. Average diameter, standard deviation and size distribution for all PS samples base on the statistics of 100 particles in TEM images.

Calculation of lattice constant (d)

Model 1: PS particles form 3D closepack colloidal crystals and the reflection originates from the interference of the incident light with (111) crystal facet. The reflection wavelength (λ) can be calculated with Equation (1), where *m* is the reflection order, *D* is the center-to-center distance between nearest spheres, n_i and V_i are, respectively, refractive index and volume fraction of each component, and ϕ is the angle between the incident light and the sample normal.

$$m\lambda = \sqrt{\frac{8}{3}} D \left(\sum_{i} n_{i}^{2} V_{i} - \sin^{2} \phi\right)^{1/2}$$
(1)
$$m\lambda = \sqrt{\frac{8}{3}} D \left(n_{PS}^{2} V_{PS} + n_{ferrofluid}^{2} V_{ferrofluid}\right)^{1/2}$$



The refractive index of water, magnetite, and PS are 1.33, 2.42, and 1.55 respectively. The refractive index of ferrofluid (Fe₃O₄% = 2%) is calculated to be 1.36. ϕ equals to 0 in our measurements, which turns Equation (1) to Equation (2). In closepack colloidal crystals, V_{ps} and V_{ferrofluid} equal to 74% and 26%, respectively. The center-to-center distance between nearest PS particles can be roughly calculated, and the results (marked in red) are summarized in the following table.

(2)

D_{PS}	λ_1	<i>D</i> calculate by λ_1	λ_2	<i>D</i> calculated by λ_2	
230	680	277			
266	742	302			
311	880	359	465	379	
348	1032	421	539	439	
397			580	473	
470			690	562	
619			920	750	

The center-to-center distance between nearest PS particles are larger than the PS diameter, which shows the PS particles do not contact to each other. Therefore, the V_{ps} and $V_{ferrofluid}$ should be decreased and increased, and the results should be recalculated accordingly. <u>Therefore, the real center-to-center distance</u> (*D*) should be slightly larger than the results listed in the above table to satisfy equation (2). The results also require the PS particles to fix or vibrate around the lattice point, if the colloid crystal expands without changing the symmetry. Model 2: PS particles form chain structures, which further aggregate to bundles, and the reflection originates from the periodic spacing between the neighbouring PS particles. The reflection wavelength (λ) can be calculated with Equation (3), where *m* is the order of diffraction, *n* is the effective refractive index of the system composed of colloids and solution, *d* is the spacing between the planes in lattice, and θ is the glancing angle between the incident light and diffraction crystal planes.

$$m\lambda = 2n_{eff}d\sin\theta \quad (3)$$

$$m\lambda = 2(n_{PS}^{2}V_{PS} + n_{ferrofluid}^{2}V_{ferrofluid})^{1/2}d$$

$$V_{PS} = \frac{4}{3}\pi(\frac{D_{PS}}{2})^{3} \div (D^{2}d) = \frac{\pi D_{PS}}{6d}$$

$$V_{ferrofluid} = 1 - \frac{\pi D_{PS}}{6d}$$

$$\left(\frac{m\lambda}{2d}\right)^{2} = 1.55^{2} \times \frac{\pi D_{PS}}{6d} + 1.36^{2} \times (1 - \frac{\pi D_{PS}}{6d})$$

$$\left(\frac{m\lambda}{2d}\right)^{2} = 0.2895 \times \frac{D_{PS}}{d} + 1.36^{2} \quad (4)$$



With θ set to 0, Equation (3) can lead to Equation (4) for practical calculation of *d*, with known *m*, λ , and *D*_{*PS*}. The results (marked in red) are summarized in the following table.

D_{PS}	ſ	d_1 calculate	λ_2	d_2 calculated	ı	d_3 calculated
	λ_{l}	by λ_1		by λ_2	Λ_3	by λ_3
230	680	233				
266	742	253				
311	880	300	465	318		
348	1032	353	539	370		
397			580	397		
470			690	472	475	488
619			920	630	632	650

The lattice constant is equal to or slightly larger than the diameter of PS particles, showing the PS particle softly contacts to each other, which is consistent with the phenomenon that increasing the external field will not blueshift the reflection wavelength anymore. Compared with Model 1, Model 2 leads to reasonable results.

Comparing the measured reflection wavelength with the calculated results

Base on the above calculations, it is known that the lattice constant is equal to or slightly larger than the diameter of PS particles, which depends on the surface repulsion and the ionic strength of the mixture. The average latticle constant (*d*) can be determined by considering all the results calculated from 1^{st} , 2^{nd} and 3^{rd} order reflection, which are listed below. As D_{PS} approximately equals to *d*, Equation 4 can be simplified to Equation 5. The results are summarized in the following table.

$$d = \frac{\sum_{i} d_{i}}{i}$$

 $\frac{m\lambda}{2d} = 1.4626 \quad (5)$

D_{PS}	Average	Measured	calculated	Measured	calculated	Measured	calculated
	d	λ_{I}	λ_1	λ_2	λ_2	λ_3	λ_3
230	233	680	682				
266	266*	742	778				
311	311*	880	910	465	455		
348	362	1032	1059	539	529		
397	397			580	581		
470	480			690	702	475	468
619	640			920	936	632	624

*the average d takes the value of particle diameter, when it is smaller than D_{PS}