

Supplementary Material (ESI) for Soft Matter

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Supplementary Information (ESI) for Soft Matter

Phase Separation of 2D Meso-scale Coulombic Crystals from Meso-scale Polarizable “Solvent”

George K. Kaufman, Samuel W. Thomas III, Meital Reches, Bryan F. Shaw, Ji Feng, and

George M. Whitesides*

Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA

12 Oxford Street, Cambridge MA, 02138

*Author to whom correspondence should be addressed:

Telephone: (617) 495-9430

Fax: (617) 495-9857

E-mail: gwhitesides@gmwgroup.harvard.edu

Experimental Details:

Materials

Teflon (T) spheres ($d_T = 3.18$ mm), Nylon-6,6 (N) spheres ($d_N = 3.18$ mm), and sheets of aluminum (0.8 and 1.0 mm) were purchased from McMaster-Carr (Princeton, NJ).

Construction of the Aluminum Frames

Circular frames were made by bending thin strips of aluminum (1.0 mm wide, 4–6 mm high, and 171–2 mm long) into circles of diameter 53.6 ± 0.2 mm. We glued the frames to square (75 mm) aluminum plates with 5-minute epoxy (Devcon), and cured the epoxy at 50 °C overnight.

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The frames enclosed similar surface areas ($2270 \pm 20 \text{ mm}^2$), which were ~ 285 times the 2D projection of the surface area of a single sphere (7.92 mm^2), and ~ 258 times the area of a hexagon that circumscribes the 2D projection of a sphere (8.73 mm^2). That is, ~ 258 spheres could hexagonally close pack in the frame.

Staining the Nylon-6,6 Spheres Blue

Nylon-6,6 spheres were immersed in an aqueous solution of Disperse Blue 14 and placed on a hot plate at a temperature of $100 \text{ }^\circ\text{C}$ for 30 min to stain the spheres blue. The blue Nylon-6,6 spheres were removed from the solution, washed with water, washed with ethanol, dried under a stream of nitrogen, and then placed in an oven at $60 \text{ }^\circ\text{C}$ for at least 2 h to remove any remaining water from the spheres.

Sputter-Coating Surfaces with Gold

The aluminum plate and frames were coated with a $\sim 220\text{-nm}$ film of gold by sputtering at 80 mA for 360 s using a sputter coater (Cressington, Model 208HR). In order to obtain a uniform coating of gold on Nylon-6,6 (Au^{N}) spheres, ~ 50 spheres were placed in a 4-cm diameter poly(styrene) Petri dish (VWR) and the dish was set at an incline on the rotating stage of the sputter coater. The spheres rolled inside the dish as the stage rotated, and were coated with a $\sim 165\text{-nm}$ (330 nm total / 2 sides of each sphere) film of gold by sputtering at 80 mA for 540 s.

To estimate the mass of the gold coating on 3.18-mm Nylon-6,6 (Au^{N}) spheres, we weighed 20 of each gold-coated and non-gold-coated sphere (N, T, Au^{N}). The gold coating on N ($18.8 \pm$

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0.2 mg) spheres weighed ~ 0.1 mg (the weight of $\text{Au}^{\text{N}} = 18.9 \pm 0.2$ mg).

Cleaning of Surfaces

To remove dirt and oil, and to discharge the spheres and the gold-coated plate prior to use, we rinsed the plate and spheres (T, N, Au^{N}) with copious amounts of water (Milli-Q, 18 M Ω) and ethanol (95%, Pharmco), and dried them with a stream of N_2 . The plate and spheres were stored in an oven at 60 °C for at least 2 h to remove any remaining water and kept in an environment with low ($8 \pm 1\%$) relative humidity during use. We measured the humidity with a hygrometer (VWR, unit 35519-050). The materials remained covered until use to prevent the accumulation of dust.

Agitation of Mixtures of Spheres

We placed mixtures of T, N and either Au^{N} spheres onto a horizontal, planar, gold-coated plate within a circular frame, and covered the frame with a gold-coated aluminum lid (Figure 1). For each experiment, the number of Au^{N} (n_{Au}) spheres equaled the sum of the numbers of T (n_{T}) and N (n_{N}) spheres, and the numbers of T and N spheres were equal ($n_{\text{Au}} = n_{\text{T}} + n_{\text{N}}$; $n_{\text{T}} = n_{\text{N}}$). The lid constrained the spheres ($d = 3.18$ mm) within a 2D space that was 4–6 mm high (Figure 1c). When the frame was too short (< 4 mm, $\sim 1.25 d$), the spheres could not move efficiently past each other on the plate. When the frame was too tall (≥ 5 mm, $1.57 d$), the spheres began to aggregate in 3D, and it became difficult to analyze images of the spheres by MATLAB (see section on Imaging and Analysis, below). We used a frame that was 4.5 mm ($1.42 d$) tall for the

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experiments in this paper so that the spheres could maintain as much mobility as possible, while remaining confined to 2D.

The plate was clamped to a square (75 mm), aluminum platform that was fixed beneath its center to a linear magnetic actuator (LinMot). We aligned the magnetic linear actuator to agitate the plate vertically (motion perpendicular to the plane spanned by the plate). The motion of the actuator and plate was approximately linear, with quasi-constant velocity (see Figure S1). Computer software controlled the amplitude and frequency of oscillation of the linear actuator, which we programmed to vibrate at frequency $\omega = 15\text{--}80$ Hz, and amplitude $A = 0.1\text{--}1.5$ mm.

Prior to agitation of the system, we discharged the spheres and plate with a ZeroStat® 3 antistatic gun (Milty). We placed a lid—a gold-coated, square (75 mm, 0.8 mm thick), aluminum sheet—on top of the frame, and secured the lid in place with c-clamps.

Shaking the system vertically (motion perpendicular to the plane of the plate) made the spheres bounce on the plate and collide with each other and the frame with random trajectories, and allowed the spheres to visit the entire area of the plate within the frame.

Imaging and Analysis: Calculation of Φ

We took photographs of the T, N and Au^N spheres with a Nikon D40 camera that was attached to a ring stand. In order to illuminate each sphere evenly (for easier image analysis), we attached a macro ring flash (SmartFlash RF46N, Phoenix) to the lens of the camera. We set the aperture to $f/25$, set the shutter speed to 1/250 s or 1/500 s, and collected the pictures with Camera Control Pro software.

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A homemade program, written in MATLAB, analyzed each photograph, and calculated the extent of separation or mixing, Φ , of the T, N and Au^N spheres in that photograph according to the following procedure. Each photograph contained n_{Au} gold-coated spheres and n_{P} polymeric spheres. To calculate Φ , we treated T and N as indistinguishable, polymeric spheres (P). We measured the position of each sphere, and calculated the distances between each pair of spheres on the plate. For each Au^N sphere, we determined how many of the 6 spheres closest to it were Au^N spheres. This number gave $n_{\text{Au-Au}}$ for *that* Au^N sphere. We then determined how many of the 6 spheres closest to that Au^N sphere were polymeric spheres (P). This number gave $n_{\text{Au-P}}$ for that Au^N sphere. We defined a “neighbor” of a sphere to be one of these six closest spheres:

$n_{\text{Au-Au}} + n_{\text{Au-P}} = n_{\text{tot}} = 6$. Figure S2a illustrates this definition.

In the event that a sphere is in a square lattice (Figure S2b), that sphere would have four spheres in its first “shell”; these spheres are of equal distance from the sphere, and for spheres of diameter (d), this distance = d . The sphere also has four spheres in its second shell (also of equal distance = $1.414 d$ from the sphere). This situation rarely occurred because the spheres rarely occupied a perfect lattice. If it were to happen, then the MATLAB program would choose the fifth and sixth spheres from the second shell at random (in the order that the program indexed the spheres—the order in which the program located the spheres from the photograph). Since the members of a square lattice were most likely to be T and N, there should be no effect on the value of Φ , since we treated T and N as indistinguishable, polymeric spheres (P).

We calculated the average of $n_{\text{Au-Au}}$ and $n_{\text{Au-P}}$ over all the gold-coated spheres to obtain the average number of Au^N spheres that neighbored each Au^N sphere, $\langle n_{\text{Au-Au}} \rangle$, and the average

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number of polymer (again, P = T or N) spheres that neighbored each Au^N sphere, $\langle n_{\text{Au-P}} \rangle$. Other

values of n_{tot} yielded quantitatively different, but qualitatively similar values and trends for Φ ,

and so we used only $n_{\text{tot}} = 6$ for all our calculations. We repeated this procedure for each P

sphere (again, we treated T and N as indistinguishable) to calculate the average number of P

spheres that neighbored each P sphere, $\langle n_{\text{P-P}} \rangle$, and the average number of Au^N spheres that

neighbored each P sphere, $\langle n_{\text{P-Au}} \rangle$. The values for $\langle n_{\text{Au-Au}} \rangle$, $\langle n_{\text{Au-P}} \rangle$, $\langle n_{\text{P-P}} \rangle$, and $\langle n_{\text{P-Au}} \rangle$ each

ranged from 0 (no neighbors of that type of sphere) to n_{tot} , and by definition obeyed Equation S1.

$$n_{\text{tot}} = \langle n_{\text{Au-Au}} \rangle + \langle n_{\text{Au-P}} \rangle = \langle n_{\text{P-P}} \rangle + \langle n_{\text{P-Au}} \rangle \quad (\text{S1})$$

To calculate the extent of separation or mixing of T, N and Au^N spheres, we calculated how many neighbors there were, on average, among spheres of the *same* phase (like neighbors), and divided this average number of like neighbors by the total number of neighbors for each sphere, according to Equations S2a and S2b.

$$\frac{\text{Number of neighbors of the same phase (Au)}}{\text{Number of neighbors of any phase (Au or P)}} = \frac{\langle n_{\text{Au-Au}} \rangle}{\langle n_{\text{Au-P}} \rangle + \langle n_{\text{Au-Au}} \rangle} \quad (\text{S2a})$$

$$\frac{\text{Number of neighbors of the same phase (P)}}{\text{Number of neighbors of any phase (Au or P)}} = \frac{\langle n_{\text{P-P}} \rangle}{\langle n_{\text{P-Au}} \rangle + \langle n_{\text{P-P}} \rangle} \quad (\text{S2b})$$

We added Equations S2a and S2b, and subtracted 1 to obtain Equation S3a. We subtracted 1 so that Φ for a random mixture would equal 0 (see next section). Combining Equations S1 and S3a yielded Equation S3b (Equation 1 in the text).

$$\Phi = \frac{\langle n_{\text{Au-Au}} \rangle}{\langle n_{\text{Au-P}} \rangle + \langle n_{\text{Au-Au}} \rangle} + \frac{\langle n_{\text{P-P}} \rangle}{\langle n_{\text{P-Au}} \rangle + \langle n_{\text{P-P}} \rangle} - 1 \quad (\text{S3a})$$

$$\Phi = \frac{\langle n_{\text{Au-Au}} \rangle}{n_{\text{tot}}} + \frac{\langle n_{\text{P-P}} \rangle}{n_{\text{tot}}} - 1 \quad (\text{S3b})$$

Limiting Behavior of Φ

Equation S3b has the limiting behavior that $\Phi = 0$ for a random mixture of spheres, or for an infinite dilution of one phase in the other. For a random mixture of spheres, $\langle n_{\text{Au-Au}} \rangle = n_{\text{tot}} p_{\text{Au}}$, where p_{Au} is the probability of having an Au–Au neighbor, and $\langle n_{\text{P-P}} \rangle = n_{\text{tot}} p_{\text{P}}$, where p_{P} is the probability of having a P–P neighbor. Equations S4a and S4b give expressions for p_{Au} and p_{P} , and Equation S4c—in which $n_{\text{S}} = n_{\text{Au}} + n_{\text{P}}$ is the number of spheres—gives Φ for a random mixture of spheres. The value of Φ reduces to zero when $n_{\text{Au}} + n_{\text{P}} \gg 2$.

$$p_{\text{Au}} = (n_{\text{Au}} - 1) / n_{\text{S}} \quad (\text{S4a})$$

$$p_{\text{P}} = (n_{\text{P}} - 1) / n_{\text{S}} \quad (\text{S4b})$$

$$\Phi = \frac{n_{\text{tot}} p_{\text{Au}}}{n_{\text{tot}}} + \frac{n_{\text{tot}} p_{\text{P}}}{n_{\text{tot}}} - 1 = \frac{n_{\text{Au}} + n_{\text{P}} - 2}{n_{\text{S}}} - 1 \approx \frac{n_{\text{S}}}{n_{\text{S}}} - 1 = 1 - 1 = 0 \quad (\text{S4c})$$

For an infinite dilution of one phase (e.g., Au^{N}) in the other (T and N), $\langle n_{\text{Au-Au}} \rangle \rightarrow 0$ and $\langle n_{\text{P-P}} \rangle \rightarrow n_{\text{tot}}$. Equation S5 shows that the value of Φ is also 0 in this limit.

$$\Phi \xrightarrow{\lim n_{\text{Au}}=0} \frac{0}{n_{\text{tot}}} + \frac{n_{\text{tot}}}{n_{\text{tot}}} - 1 = \frac{n_{\text{tot}}}{n_{\text{tot}}} - 1 = 1 - 1 = 0 \quad (\text{S5})$$

Equation S3b also has the limit that $\Phi = 1$ for completely separated phases (Figure S2c). In this

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limit, $\langle n_{\text{Au-P}} \rangle = \langle n_{\text{P-Au}} \rangle = 0$, so by Equation S1, $\langle n_{\text{Au-Au}} \rangle = \langle n_{\text{P-P}} \rangle = n_{\text{tot}}$.

$$\Phi \xrightarrow{\text{lim non-touching}} \frac{n_{\text{tot}}}{n_{\text{tot}}} + \frac{n_{\text{tot}}}{n_{\text{tot}}} - 1 = 1 + 1 - 1 = 1 \quad (\text{S6})$$

As mentioned in the text, $\Phi < 0$ (about -0.20) for mixtures of spheres in which we intentionally placed T and N spheres between pairs of Au^{N} spheres, as in Figure 2 (left) or Figure S2a (top).

To generate center-surround (core-shell) configurations of spheres, we drew blue circles (to represent Nylon spheres), light blue circles (to represent Teflon spheres), and red circles (to represent gold-coated spheres). We positioned these circles in the configurations shown in Figure S3 using Adobe Illustrator CS2, and then calculated Φ for each configuration (as described above) to obtain values of $\Phi_{\text{max}}^{\text{C-S}}$. Dividing Φ , calculated for our experimentally obtained configurations of spheres, by the appropriate value of $\Phi_{\text{max}}^{\text{C-S}}$ (same number of T, N, and Au^{N} spheres) gave the values of Φ^* reported in the text.

Effect of Humidity on Separation

Agitation was performed in a glove bag (Captair Pyramid; Erlab, Inc.). We measured the humidity and temperature with a hygrometer (VWR, unit 35519-050). To obtain 8% RH at room temperature (21 ± 2 °C), we flowed “dry” house air, that we did not treat further, in to the glove bag. To obtain $\text{RH} \approx 20\text{--}30\%$, we flowed this “dry” air via a diffuser through water that was in a sealed Erlenmeyer flask to obtain water-saturated air, which we combined with “dry” air. We adjusted the rates of flow of dry air and water-saturated air that entered the glove bag until the RH inside it was 20–30%. Experiments in which we agitated T, N, and Au^{N} ($\rho = 0.62$ and 0.70)

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for 60 min (5-min intervals at $A = 0.60$ mm, $\omega = 60$ Hz) showed that separation was not

quantifiably different between RH = 8% and 20–30%. For all experiments in this paper, the relative humidity in the glove bag was $8 \pm 1\%$ at room temperature.

Measurement of the Charge on Spheres

For measuring the charge on T, N, and Au^N spheres, we fabricated a Faraday cup out of three concentric (inner, middle, and outer) aluminum boxes (Figure S4a). Each box had an ~1.25-mm circular hole cut into its lid so that spheres could pass into the interior of the inner box without touching the middle or outer boxes. We connected the boxes to an electrometer that measured the charge (as a function of time) on the inner box with respect to the middle box, and held the outer box at ground. A homemade program in LabView recorded the charge measured by the electrometer. The charge on a sphere corresponded to the difference in charge inside the cup before and after the sphere entered the cup (Figure S4b–d).

We measured the charge on each sphere by removing it individually from the plate with poly(butylene terephthalate) (PBTP; Table S1), stainless-steel, or wooden needle-nose tweezers, and dropping it into the Faraday cup. The charge determined by the Faraday cup depended on the material of the tweezers used to hold the sphere (Tables S2 and S3). The stainless-steel tweezers appeared to discharge the spheres: the charge of T and N spheres measured with stainless-steel tweezers was ~60–80% of their charge with either PBTP or wooden tweezers. The charges measured with PBTP and wooden tweezers were also slightly different for reasons beyond the scope of this study. The differences were not consistent with the spheres

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tribocharging against the tweezers, since the charges of neutralized T and N spheres did not differ when using PBTP or wooden tweezers. For consistency, we used PBTP tweezers for all measurements reported in this communication.

In order to determine the effect of the electrostatic environment on the charge on T, N, and Au^N spheres, we agitated 50 T, 50 N, and 100 Au^N spheres for 3 min at $\omega = 60$ Hz, and $A = 0.26$ mm. We removed from the plate all the spheres of a first kind (e.g., T) to measure their charge, then all spheres of a second kind (e.g., N), and then all the spheres of the third kind (e.g., Au^N).

To determine the kinetics of charging of T, N, and Au^N spheres, we agitated two densities of spheres ($\rho = 0.77$, $n_{\text{Au}} = 100$, $n_{\text{N}} = n_{\text{T}} = 50$; and $\rho = 0.46$, $n_{\text{Au}} = 60$, $n_{\text{N}} = n_{\text{T}} = 30$) on the plate at $A = 0.26$ mm, and $\omega = 60$ Hz for intervals of $t = 0, 3, 5, 7.5, 10, 15, 30, 60$, and 180 s. Prior to agitating the spheres, we neutralized the charge on the spheres with a Zerostat gun. After each interval of agitation, we measured the charge on the Au^N spheres, then N, and then T, with PBTP tweezers, as described above. Figure S5a shows that the magnitude of the average charge on N and T increased linearly, at similar rates, before reaching constant values at ~ 10 s ($\rho = 0.77$) or ~ 30 s ($\rho = 0.46$).

To determine how long it took for charge to dissipate spontaneously (i.e., by lateral movement of charge among the beads, rather than by being discharged with a Zerostat gun), we measured the charge as a function of time on a mixture of stationary spheres that had been agitated for 3 min at $A = 0.26$ mm, and $\omega = 60$ Hz (Figure S5b). The charge on T and Au^N spheres did not change significantly over the course of 68 h. Over this time, the charge on N decreased by ~ 55 pC (11%), which was slightly larger than the standard deviation of charges

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measured on N at any given time. We conclude that, during a typical 1–60 min experiment,

there is no significant dissipation of charge from any of the spheres.

Effect of Amplitude on Separation

To determine if the amplitude (A) affected the separation of T and N from Au^N, we agitated 40 T, 40 N, and 80 Au^N spheres at $\omega = 60$ Hz and a range of amplitudes ($A = 0.25$ – 1.02 mm).

Figure S6 shows that there was a slight increase in separation (Φ^*) with increasing A at low agitation ($A = 0.25$ – 0.60 mm), but no clear trend at high agitation ($A = 0.66$ – 1.02 mm).

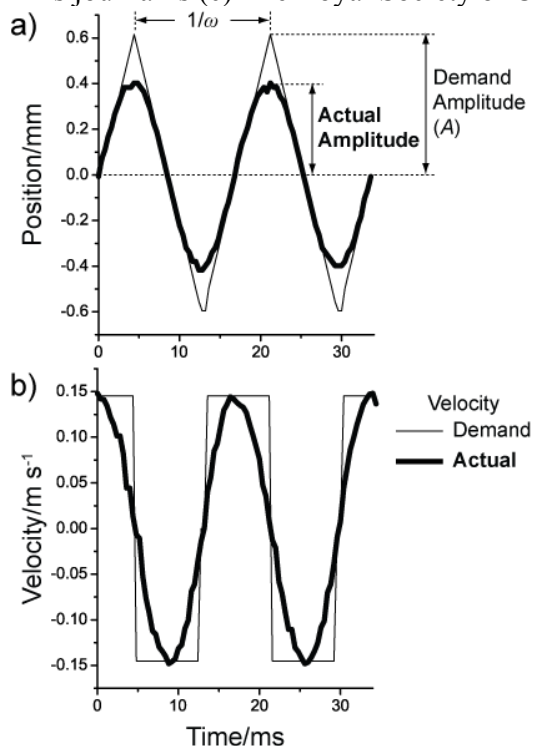


Figure S1. a) Position and b) velocity of the dish, perpendicular to the plane spanned by the dish, for agitation of the spheres, dish, and lid at $\omega = 60$ Hz, $A = 0.60$ mm. a) We programmed the linear magnetic motor to oscillate in a triangular wave, in which the amplitude, A , was half the difference between the maximal and minimal vertical positions of the dish, and the frequency, ω , was the inverse of the time between maxima of the wave. The amplitude of the motor (Actual amplitude) deviated from the programmed (Demand) position at the maxima and minima, where the acceleration of the system would have had to exceed the maximal allowed acceleration of the motor to achieve the programmed wave function. b) The actual maximal speed of the motor was, on average, within $\sim 2\%$ of the programmed (Demand) maximal speed (144 mm s^{-1}).

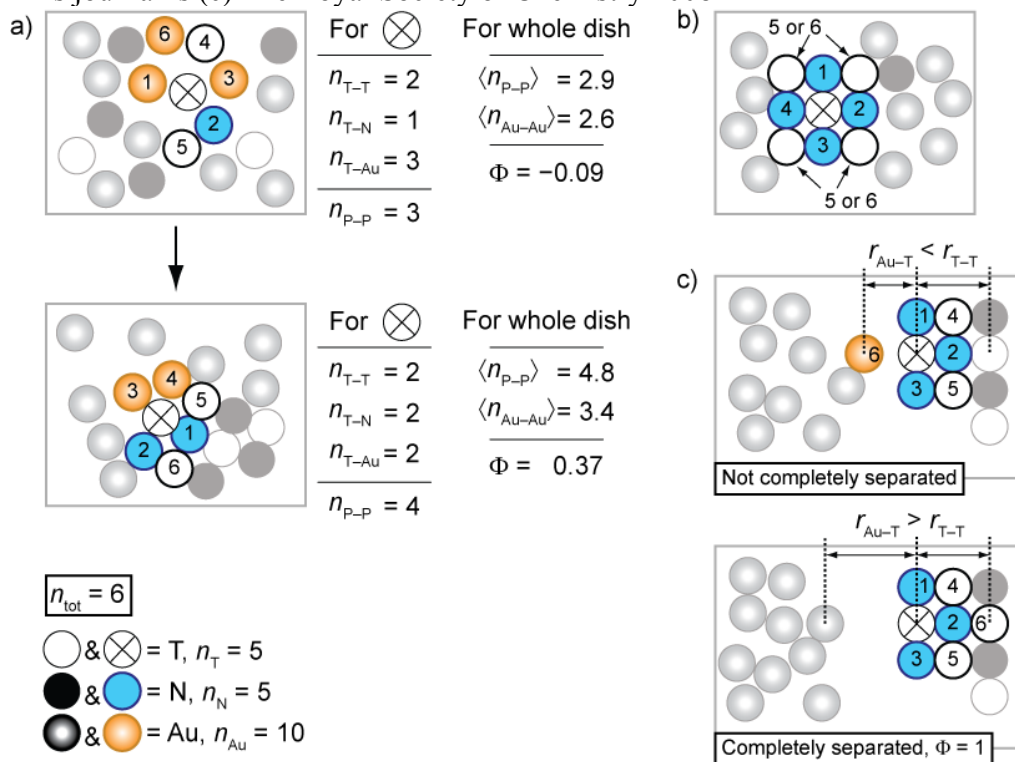


Figure S2. a) Illustration of the six neighbors ($n_{tot} = 6$) of a Teflon sphere (marked with an \times) for the two configurations of spheres shown in Figure 1a. The six neighbors are numbered in order of their proximity to the marked Teflon sphere. The values for the “whole dish” are averages over all 10 polymeric spheres, $\langle n_{P-P} \rangle$, and all 10 gold-coated spheres, $\langle n_{Au-Au} \rangle$, shown in these configurations. We use these average values to calculate Φ according to Equation 1. b) T and N in a square lattice; four N spheres (blue) occupy the first “shell” around the marked T sphere, and four T spheres (white) occupy the second “shell”.

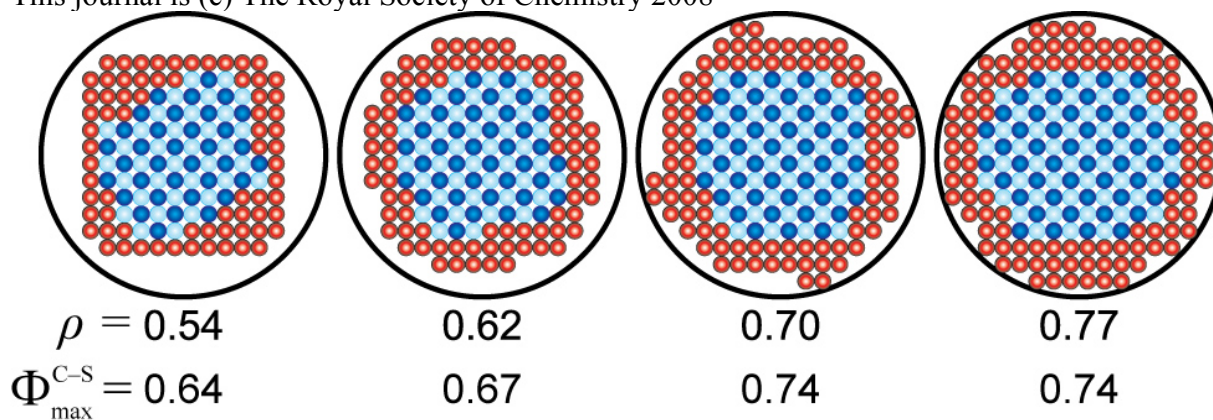


Figure S3. Center-surround (C-S) configurations of spheres in which T (white) and N (blue) spheres occupy the center and Au (red) spheres occupy the perimeter—the surround—of each configuration. We used eqn (1) to calculate the value of Φ for each configuration; the value of $\Phi_{\max}^{\text{C-S}}$ at each density of spheres (ρ) is defined this value of Φ .

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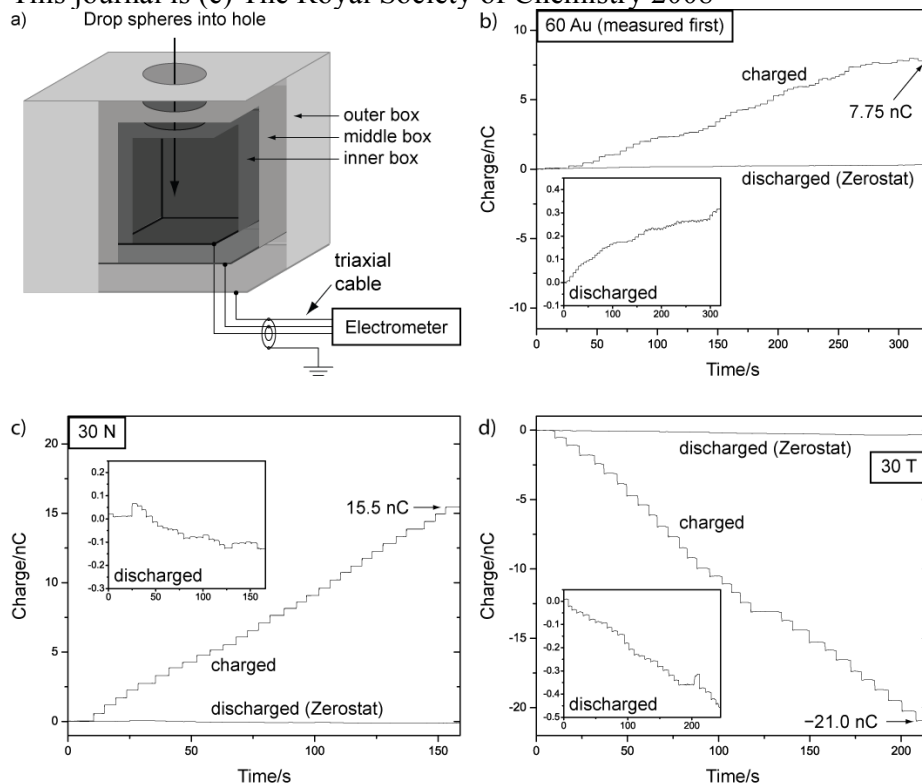


Figure S4. a) Schematic of the Faraday cup. We measured the charge on each sphere by removing it from the surface with poly(butylene terephthalate) (PBTP) tweezers, and dropping it into the Faraday cup, which was connected to an electrometer that measured charge as a function of time. The integration time on the electrometer was set to 16.7 ms, and the electrometer recorded the value of the charge at 57-ms intervals. We used this cup to measure the charge on (b) 60 Au^N spheres (measured, one at a time, “first”; that is, while the N and T remained in the dish), (c) 30 N spheres, added one at a time, and (d) 30 T spheres, added one at a time, immediately after agitating the spheres ($\omega = 60$ Hz, $A = 0.26$ mm) for 3 min. The average charges on these spheres were +129 pC (Au^N), +517 pC (N), and -700 pC (T), similar to those charges shown in Figure 3d, and those reported in Table S1. Insets (b–d): Charge on spheres that

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were discharged (their electrostatic charge was neutralized) with a Zerostat gun.

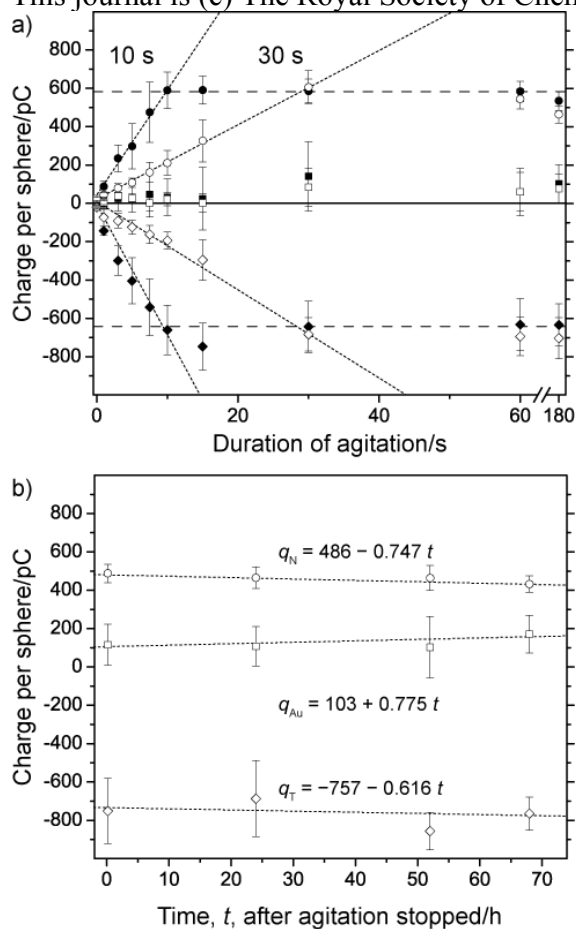


Figure S5. Average charge on Teflon (q_T , diamonds), Nylon-6,6 (q_N , circles) and gold-coated (q_{Au} , squares) spheres. a) Charge as a function of the duration of agitation at $A = 0.26$ mm and $\omega = 60$ Hz on an ungrounded dish. The magnitude of the charges on N and T increased linearly (dotted lines), and at similar rates, before reaching a plateau (dashed lines) at 10 s (closed symbols; 50 T, 50 N, and 100 Au^N) or 30 s (open symbols; 30 T, 30 N, and 60 Au^N). b) Charge as a function of time after agitation ($A = 0.26$ mm, $\omega = 60$ Hz, 3 min) had stopped. We measured the charge that remained on the spheres after sitting without agitation for 10 min, 24 h, 52 h, and 68 h. The dotted lines are weighted linear fits to the data. These charges did not differ

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significantly when we used a grounded dish.

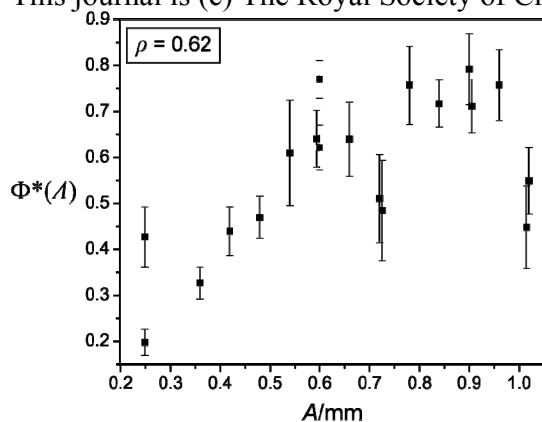


Figure S6. The value of $\Phi^*(A) = \Phi(A)/\Phi_{\text{max}}^{\text{C-S}}$ for the separation of 40 T, 40 N, and 80 Au^N spheres ($\rho = 0.62$) as a function of the amplitude (A) of oscillation of the plate. We agitated the spheres for 15 min—in 30-s intervals—at $\omega = 60$ Hz (as in Figure 4). The reported values are the average of the values of $\Phi^*(A)$ over the last 10 min of each trial; the lengths of the error bars represent the standard deviation obtained from these values. For clarity, the values of A are offset for the multiple trials at $A = 0.60$, 0.72 , and 1.02 mm. The maximal theoretical value for Φ^* is 1.0.

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Table S1. Charge measured on spheres after agitation of spheres at $A = 0.26$ mm, $\omega = 60$ Hz for

3 min on a grounded plate.^a

Order measured			Charge, q /pC		
1	2	3	q_T	q_N	q_{Au}
Au			—	—	24 ± 7
N			—	470 ± 98	—
T			-383 ± 133	—	—
Au	N		—	638 ± 56	-335 ± 95
N	Au		—	633 ± 49	29 ± 13
Au	T		-658 ± 91	—	487 ± 96
T	Au		-669 ± 58	—	9 ± 5
N	T		-764 ± 94	651 ± 47	—
T	N		-803 ± 48	593 ± 64	—
Au ^b	N	T	-718 ± 111	537 ± 54	85 ± 112
Au ^b	T	N	-829 ± 80	570 ± 40	144 ± 145
N	Au ^c	T	-732 ± 87	594 ± 53	388 ± 109
N	T	Au ^e	-739 ± 89	634 ± 47	9 ± 25
T	Au ^d	N	-811 ± 66	539 ± 53	-292 ± 94
T	N	Au ^e	-785 ± 66	587 ± 41	32 ± 17
Discharged spheres:			-16 ± 11	-2 ± 13	8 ± 10

^a We used PBTP tweezers because the metal tweezers partially discharges the spheres (see

Tables S2 and S3). ^b Corresponds to Figure 3a. ^c Corresponds to Figure 3b. ^d Corresponds to

Figure 3c. ^e Corresponds to Figure 3d.

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Table S2. Charge measured on spheres using PBTP, wood, and metal tweezers.^a

Material of Tweezers	Charge, q/pC		
	q_T	q_N	q_{Au}
PBTP	-718 ± 111	537 ± 54	85 ± 112
Wood	-826 ± 62	539 ± 45	-66 ± 90
Metal	-527 ± 134	419 ± 12	0 ± 0
Discharged spheres ^b			
PBTP	-15 ± 6	-1 ± 7	45 ± 13
Wood	-17 ± 7	-5 ± 8	-68 ± 10
Metal	-33 ± 17	7 ± 6	0 ± 0

^a Agitated 50 T, 50 N, and 100 Au^N spheres at $A = 0.26$ mm, $\omega = 60$ Hz for 3 min on a grounded dish, and measured the charge on 25 Au^N, then 25 N, then 25 T spheres. ^b Charge on spheres that were discharged (their electrostatic charge was neutralized) with a Zerostat gun.

Table S3. Charge measured on spheres using PBTP and metal tweezers.^a

Material and Order of Tweezers	Charge, q/pC	
	q_T	q_N
PBTP	-699 ± 122	675 ± 53
Metal, PBTP ^b	-743 ± 121	624 ± 145
Metal	-428 ± 103	459 ± 39
PBTP, Metal ^c	-418 ± 91	447 ± 48

^a Agitated 50 T, 50 N, and 100 Au^N spheres at $A = 0.26$ mm, $\omega = 60$ Hz for 30 s on a grounded dish, and measured the charge on 25 Au^N, then 25 N, then 25 T spheres. ^b “Metal, PBTP” = removed each sphere from the dish with metal tweezers, and transferred the sphere to PBTP tweezers before dropping the sphere into the Faraday cup. ^c “PBTP, Metal” = removed each sphere from the dish with PBTP tweezers, and transferred the sphere to metal tweezers before dropping the sphere into the Faraday cup.