Supplemental Materials: Universal reshaping of arrested colloidal gels via active doping

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DETAILS FOR PATCHY PAIR POTENTIALS

To describe the dynamics of square lattice forming particles, we adopted a patchy particles model from Ref. [1]. In this model, each particle is modeled as a sphere of diameter σ with four distinct attractive regions (patches) placed orthogonal to each other as shown in green in Fig 1. Each particle undergoes Brownian dynamics at a constant temperature T according to the translational and rotational equations of motion introduced in Eqs. 1. and 2 in the main text.

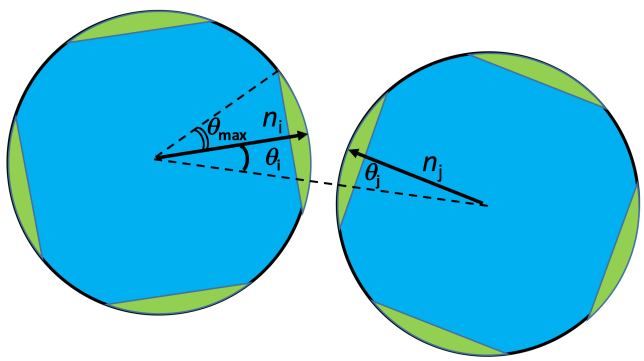

FIG. 1. Illustration showing our four patch model. The hydrophobic patches are shown in blue, whereas the hydrophilic region is depicted in yellow.

The interaction potential, V_{ij}, between any two patchy particles has the form

$$V_{ij}\left(r_{ij}, \theta_i, \theta_j\right) = V_{\mathrm{R}}(r_{ij}) + V_{\mathrm{A}}(r_{ij})\phi(\theta_i)\phi(\theta_j) \tag{1}$$

where

$$V_{\mathrm{R}}(r_{ij}) = \begin{cases} 4\varepsilon_{rep}\left(\dfrac{\sigma}{r_{ij}}\right)^{12} & r_{ij} \leq 1.5\sigma \\ 0 & r_{ij} > 1.5\sigma \end{cases} \tag{2}$$

accounts for the excluded volume interactions between the particles, and

$$V_{\mathrm{A}}(r_{ij}) = 4\varepsilon\left[\left(\frac{\sigma/2}{r_s + \frac{\sigma}{2}\times 2^{1/6}}\right)^{12} - \left(\frac{\sigma/2}{r_s + \frac{\sigma}{2}\times 2^{1/6}}\right)^{6}\right] \tag{3}$$

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which is also cut off at 1.5σ, accounts for their possible angular attraction. Here ε is the binding energy, r_{ij} is the distance between the centers of particles i and j, while $r_s = |r_{ij} - \sigma|$ is the distance between the surfaces of the particles. The angular potential $\phi(\theta_i)$, setting the interaction between any two hydrophobic patches, is given by

$$\phi(\theta_i) = \begin{cases} 1 & \theta_i \leq \theta_{max} \\ \cos^2\left(\dfrac{\pi(\theta_i - \theta_{max})}{2\theta_{tail}}\right) & \theta_{max} \leq \theta_i \leq \theta_{max} + \theta_{tail} \\ 0 & \text{otherwise} \end{cases} \tag{4}$$

where θ_i is defined as the angle between an hydrophobic patch unit vector $\boldsymbol{n}_i$ and the inter-particle vector $\boldsymbol{r}_{ji} = \boldsymbol{r}_j - \boldsymbol{r}_i$ (similarly θ_j is the angle between patch vector $\boldsymbol{n}_j$ and inter-particle vector $\boldsymbol{r}_{ij} = -\boldsymbol{r}_{ji}$). The corresponding expression for $\phi(\theta_j)$ is given by Eq. 4. The angular potential ϕ is a smooth step function that modulates the angular dependence of the potential and is equal to 1 within the region $\theta_i < \theta_{max}$ and decays to zero following the expression above. We selected $\theta_{max} = 18°$, and $\theta_{tail} = 20°$ to generate a sufficiently smooth potential at the Janus interfaces. For the triblock Janus colloid, the two hydrophobic patches are located at opposite poles of the colloid with each hydrophobic patch accounting for $\approx 28\%$ of the particle surface area. The polar equatorial region accounts for the remaining 44% of the surface area. Here, $\theta_{max} = 25.2°$, and $\theta_{tail} = 25°$ which is identical to the potential implemented in Ref. [2]. This flavor of potential was adapted from the model introduced in Ref. [3], and is appropriate to describe the behavior of patchy particles at high salt concentration, where the charge on the polar side of the colloid is well screened and short ranged.

SUPPLEMENTAL VIDEOS

S1. Isotropically attractive passive colloids (blue) in two dimensions with interaction energy $\varepsilon/k_bT = 5$ in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.3$ with an active doping fraction of $\chi = 0.1$ and a self-propelling speed of $U = 50$. Self-propulsion is introduced once the gel has been allowed to coarsen and the video shows the evolution of the system from the kinetically frustrated extended gel to the microphase of monodisperse crystallites.

S2. Isotropically attractive passive colloids (blue) in two dimensions with interaction energy $\varepsilon/k_BT = 5$ in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.3$ with an active doping fraction of $\chi = 0.1$ and a self-propelling speed of $U = 50$. Self-propulsion is introduced once the gel has been allowed to coarsen and the video shows the evolution of the system from the kinetically frustrated extended gel to the microphase of monodisperse crystallites. This video highlights the novel shielding mechanism and the dynamic layering that takes place at the surface of a passive cluster.

S3. Triblock Janus colloids with interaction energy $\varepsilon/k_BT = 15$ (blue), designed to form a kagome lattice in two dimensions, in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.2$ with an active doping fraction of $\chi = 0.016$ and a self-propelling speed of $U = 0$ (equilibrium system). This video illustrates the dynamics of the kinetically frustrated gel starting from a homogeneous initial configuration.

S4. Triblock Janus colloids with interaction energy $\varepsilon/k_BT = 15$ (blue), designed to form a kagome lattice in two dimensions, in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.2$ with an active doping fraction of $\chi = 0.016$ and a self-propelling speed of $U = 60$. This video illustrates the dynamics of the microphase of monodisperse crystallites starting from a homogeneous initial configuration.

S5. Four-patch colloids (blue) with interaction energy $\varepsilon/k_BT = 20$, designed to form a square lattice in two dimensions, in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.2$ with an active doping fraction of $\chi = 0.016$ and a self-propelling speed of $U = 0$ (equilibrium system). This video illustrates the dynamics of the kinetically frustrated gel starting from a homogeneous initial configuration.

S6. Four-patch colloids (blue) with interaction energy $\varepsilon/k_BT = 20$, designed to form a square lattice in two dimensions, in the presence of purely-repulsive active colloids (orange). The overall density is $\phi = 0.2$ with an active doping fraction of $\chi = 0.016$ and a self-propelling speed of $U = 50$. This video illustrates the dynamics of the microphase of monodisperse crystallites starting from a homogeneous initial configuration.

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[3] W. L. Miller and A. Cacciuto, Hierarchical self-assembly of asymmetric amphiphatic spherical colloidal particles, Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys. **80**, 021404 (2009).