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Elasticity-mediated morphogenesis in interfacial colloidal assemblies during droplet evaporation

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Abstract: We study the self-assembly of colloidal microgel particles at the quasi-two-dimensional air-water interface of a drying droplet. Using bright-field microscopy, we demonstrate that increasing particle elasticity drives interfacial organization from repulsion-stabilized crystallization to attraction-dominated gelation, via diverse metastable structures including clusters, voids and anisotropic aggregates. Molecular dynamics simulations, using an effective potential that captures the interplay between hydrophobic, capillary, steric and dipolar interactions, reproduce the overall phenomenology of the observed colloidal morphogenesis. Our findings establish particle elasticity as a key parameter governing non-equilibrium structural organization, phase separation and pattern formation of colloids at an interface.

Keywords: colloids; elasticity; droplet evaporation; self-assembly; microscopy; effective interaction

1 Introduction

Non-equilibrium structural organization, a key feature in biological and soft-matter, drives phase transitions and the

formation of complex morphologies across different spatiotemporal scales [1], [2]. While rigid colloids have provided significant insights into two-dimensional (2D) phase transitions and interfacial self-assembly [3]–[5], they fail to capture the structural evolution exhibited by deformable colloids. In this case, internal degrees of freedom and particle elasticity can drastically modulate interparticle interactions, and therefore the emergent suspension properties [6]–[9]. Due to their core-corona architecture [10], polymer-based soft colloidal microgels have recently been employed to study how particle deformability influences non-equilibrium structural organization in glassy materials [7], non-Newtonian flows [11] and crystallization [12], [13].

Microgels spontaneously adsorb at fluid interfaces due to their amphiphilic nature, while also deforming to reduce interfacial tension [9], [14], [15]. During the drying of sessile droplets, these adsorbed microgels inhibit the coffee ring effect [16] and form various 2D structures dictated by local physicochemical conditions [15], [17]. Beyond its fundamental role in probing phase transitions and the organization of particles into higher order complex structures [4], [14], [18]–[21], interface-assisted self-assembly of colloidal particles is utilized for stabilizing foams and emulsions [22], [23], and in nanolithographic applications [8]. Although the role of particle elasticity in pattern formation by microgel deposits has been reported [24], its influence on interparticle interactions and the resulting dynamics of the self-assembled architectures remains largely unexplored.

In this letter, we utilize the air-water interface of evaporating sessile droplets as a simple platform to capture elasticity-mediated changes in effective interactions and colloidal morphogenesis. We demonstrate that changes in particle elasticity drive a series of distinct morphological crossovers in the non-equilibrium structural organization of poly(*N*-isopropylacrylamide) (PNIPAM) microgels [25], [26]. By drying sessile droplets of dilute suspensions, we harnessed spatial gradients in microgel concentration at the droplet air-water interface to study self-assembly over a range of local particle area fractions, ϕ . We report that soft, highly deformable microgels transform from isolated circular clusters to foam-like void structures with

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increase in ϕ , eventually stabilizing into ordered, faceted hexagonal lattices at later times. In contrast, increasing microgel crosslinker density enhances particle elasticity, promoting anisotropic structures such as chain-like aggregates that percolate into 2D disordered gel networks at high ϕ . We quantify the observed morphological diversity through the global bond orientational order parameter, $\langle |\psi_6| \rangle$, and pair correlation function, $g(r)$. We propose an effective interparticle potential, $U(r)$, incorporating hydrophobic, steric, capillary and electrostatic (dipolar) contributions [27]–[30]. Altering the steric range and dipolar strength in molecular dynamics (MD) simulations allows us to replicate the entire array of experimental morphologies. In summary, microgel elasticity drives a crossover from ordered hexagonal lattices to disordered gel-like networks due to modulation in the interparticle potential. Our study provides a unified picture of interfacial morphogenesis in deformable colloidal systems and enables the tunable design of complex 2D structures.

2 Materials and methods

PNIPAM microgels of different crosslinker densities were synthesized [31] (Supplementary Material, ST1). Scanning electron microscopy (SEM) images in Figure 1(a) display microgels synthesized over a range of crosslinker densities. Height profiles in Figure 1(b), measured using atomic force spectroscopy (AFM), show that a lower crosslinker density leads to reduced heights of the dried microgels, indicating higher deformability [24]. Figure 1(c) shows that the swelling ratio α [11], the ratio of mean hydrodynamic diameters of the microgels in the maximally swollen (20 °C) and collapsed (45 °C) states (Supplementary Material, ST2), decreases with increasing crosslinker (N,N'-Methylenebis(acrylamide), MBA) content. This reduction in α implies a higher elastic modulus, consistent with the behavior of stiffer microgels. In the rest of the letter, we have used $\frac{1}{\alpha}$ as a measure of particle elasticity.

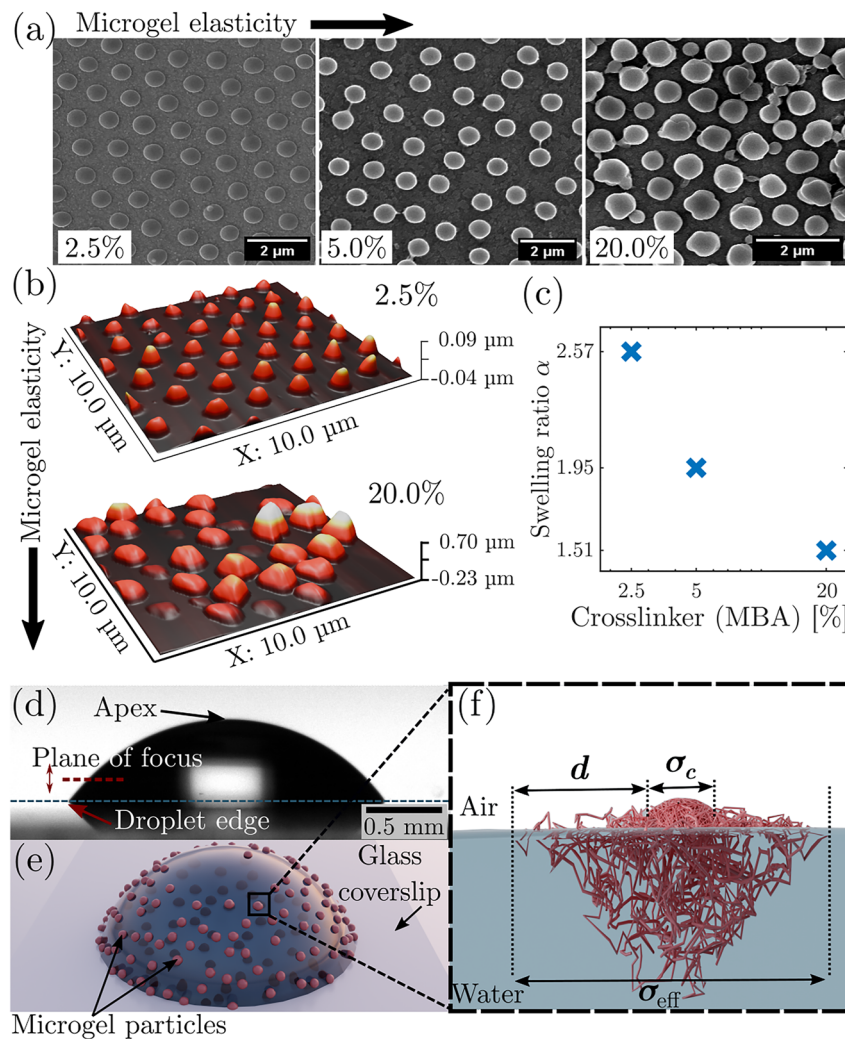


Figure 1: Microgel characterization and experimental setup. (a) SEM images of dried microgels of different crosslinker (MBA) densities, 2.5 %, 5 % and 20 %. (b) AFM height profiles of dried microgels of two different crosslinker densities. (c) Swelling ratio α versus crosslinker density. (d) Side-view micrograph of a sessile droplet on a glass substrate. (e) Schematic illustration of the droplet and microgel particles (purple spheres) adsorbed at the air-water interface. (f) Zoomed-in view of a single adsorbed microgel.

We dried 1 μL sessile droplets of microgel suspensions (particle concentrations $\sim 0.002\text{--}0.008\%$ w/v) on glass coverslips under ambient conditions (temperature = $22 \pm 2^\circ\text{C}$ and relative humidity $\approx 40\text{--}45\%$). A side-view micrograph of a droplet, imaged using a contact angle goniometer, is shown in Figure 1(d) and a schematic of the same is shown in Figure 1(e). Owing to their core-corona architecture, the microgels flatten into a fried egg configuration (side-view schematic in Figure 1(f)) to minimize interfacial energy. The diameter of the incompressible core σ_c and thickness of the compressible corona d are labeled in Figure 1(f). Microgel self-assembly at the air-water interface during droplet evaporation (Supplementary Material, Figure S3) was monitored in bright-field transmission mode with a 63X oil-immersion

objective. These images were further processed using Ilastik [32] and MATLAB (Supplementary Material, Figure S4).

3 Experimental results

3.1 Observation of evaporation-induced self-assembly at an interface

In evaporating sessile droplets, microgel area fraction (ϕ) at the air-water interface increases via two synergistic mechanisms. Capillary flows transport the colloids toward the droplet edge (triple phase contact line, TPCL) [16], leading to a gradual increase in ϕ as the TPCL is approached. Simultaneously, ϕ increases due to evaporation-induced water loss

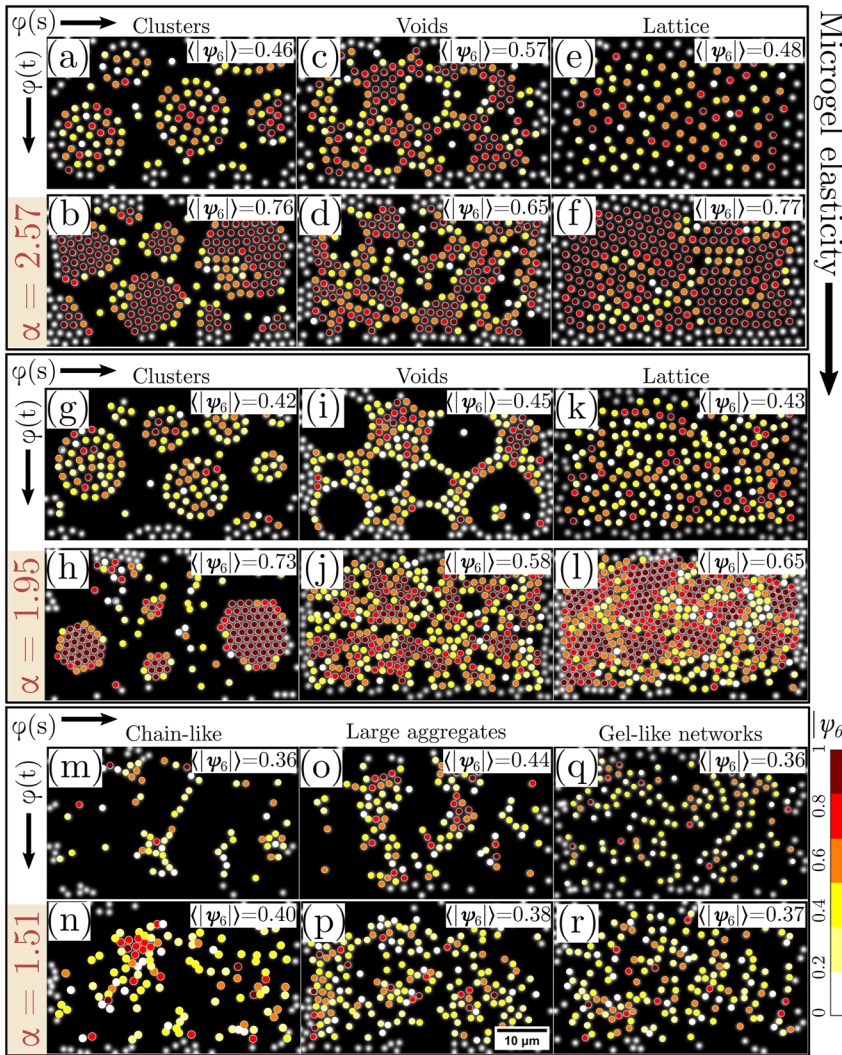


Figure 2: Reconstructed bright-field micrographs (color-coded by $|\psi_6|$) showing the spatiotemporal self-assembly of microgel cores for three particle elasticity values (top to bottom panels). Top panel (a–f): soft microgels ($\alpha = 2.57$). Middle panel (g–l): microgels of intermediate elasticity ($\alpha = 1.95$). Bottom panel (m–r): stiff microgels ($\alpha = 1.51$). The local area fraction (ϕ , denoted by $\phi(s)$ and $\phi(t)$) increases from left to right and top to bottom respectively.

and migration of microgels from the bulk. In our experiments, the spatial variation of ϕ , denoted by $\phi(s)$, was significantly more pronounced than its temporal increase, $\phi(t)$. This allowed us to investigate a wide range of distinct assembly processes at the interface of the evaporating droplet. Diverse metastable self-assembled structures were observed to coexist and evolve spatiotemporally, driven by microgel rearrangements and the increase in ϕ . Representative reconstructed micrographs of self-assembled microgels of different elasticities are displayed in Figure 2, with corresponding raw images provided in Figure S5 of the Supplementary Material. The microgels are color-coded by local bond orientational order $|\psi_6|$. The corresponding distributions are presented in Figure S6 of the Supplementary Material.

3.1.1 Soft microgel self-assembly

Soft microgels ($\alpha = 2.57$, Figure 1(b) and (c)), characterized by low particle elasticity, assembled into ordered circular clusters ($\langle |\psi_6| \rangle = 0.46$) at low ϕ (Figure 2(a) and Supplementary Material Movie, SV1). During the course of droplet evaporation, these circular clusters transformed to faceted structures ($\langle |\psi_6| \rangle = 0.76$), displayed in Figure 2(b). Time-lapse images of cluster coalescence are displayed in Figure S7 of the Supplementary Material. With increase in ϕ as the TPCL was approached, 2D foam-like structures with circular voids surrounded by microgel walls were observed (Figure 2(c) and Supplementary Material Movie, SV2). Interestingly, circular voids coalesced to form bigger voids (Supplementary Material, Figure S8) and progressively adopted non-circular shapes as the microgels ordered with elapsed time, as reflected by the increase in $\langle |\psi_6| \rangle$ from Figure 2(c) to Figure 2(d). At the highest ϕ , the soft microgel particles self-assembled into loosely arranged hexagonal lattices (Figure 2(e) and (f) and Supplementary Material Movie SV3), with $\langle |\psi_6| \rangle \approx 0.77$ in Figure 2(f). The observed monotonic increase in $\langle |\psi_6| \rangle$ with ϕ indicates a systematic crossover to more ordered assemblies.

3.1.2 Self-assembly of microgels of intermediate elasticity

At early times, microgels of intermediate elasticity ($\alpha = 1.95$) exhibited self-assembled structures (Figure 2(g)–(i)) that were qualitatively similar to the softer counterparts. As the TPCL was approached, the structures became relatively disordered (Figure 2(k)), but progressively regained order at late times, as evidenced from Figure 2(h), (j) and (l), where $\langle |\psi_6| \rangle = 0.58$ – 0.73 . This intermediate regime also exhibited a rich variety of configurations, including microgel

clusters coexisting with chain-like aggregates, as well as highly ordered domains alongside disordered assemblies and 2D foams (Supplementary Material, Figure S9 and Supplementary Material Movies SV4(a) and SV4(b)).

3.1.3 Stiff microgel self-assembly

Highly elastic microgel particles ($\alpha = 1.51$) exhibited markedly different self-assembly pathways. Rather than forming ordered clusters, they assembled into chain-like aggregates at low ϕ (Figure 2(m), $\langle |\psi_6| \rangle = 0.36$). As shown in Figure S10 of the Supplementary Material, the self-assembled morphologies evolved by folding [2] and interconnection with neighboring aggregates (Figure 2(n) and Supplementary Material Movie SV5). With increase in ϕ , the chains transformed into anisotropic clusters (Figure 2(o)) that eventually percolated into disordered 2D gel-like networks (Figure 2(p), (q), and (r) and Supplementary Material Movie, SV6). These structures are reminiscent of those observed in simulations of strongly attractive colloids [33].

4 MD simulations

Several studies have explored the emergence of complex colloidal mesostructures, such as foams, cellular structures, ordered aggregates and voids at interfaces [34], [35] and explained the results in terms of inter-particle interactions [36], [37]. Contact line undulations arising from surface roughness or chemical inhomogeneity of the adsorbed microgels result in interparticle capillary attractions [37]. In addition, an effective steric repulsion emerges from the compression of the polymer brushes that constitute the microgel coronas [6]. The unfavorable interaction between polymer moieties and the aqueous medium further leads to a purely entropic, short-range hydrophobic attraction between microgel cores [27]. Moreover, microgels at fluid interfaces behave as dipoles due to the asymmetric screening of surface charges at the interface, giving rise to long-range repulsive interactions [30].

To elucidate the contributions of these competing interactions in driving the observed self-assembled morphologies [38], we propose an effective pair potential, $U(r)$, as a function of center-to-center microgel separation, r :

$$U(r) = \begin{cases} \infty, & r \leq \sigma_c, \\ U_h + U_s + U_c + U_d, & \sigma_c < r < 8\sigma_c, \\ 0, & r \geq 8\sigma_c \end{cases} \quad (1)$$

In the above expression, the hard wall repulsion at $r \leq \sigma_c$ arises from volume exclusion of the microgel cores. The different contributions to $U(r)$ for $\sigma_c < r < 8\sigma_c$ are: (i) *Hydrophobic attraction*: $U_h = -W_0 \exp[-(h/\lambda_0)]$ [27], where W_0 and λ_0 denote the strength and range of the interaction, and $h = r - \sigma_c$ is the surface separation between two microgel cores. (ii) *Steric repulsion*: A two-dimensional Hertzian expression results in $U_s = S\sigma_{\text{eff}}^2 \left(1 - \frac{r}{\sigma_{\text{eff}}}\right)^2 \Theta(\sigma_{\text{eff}} - r)$ [6], where S is the steric repulsion strength, $\sigma_{\text{eff}} = \sigma_c + 2d$ is the effective particle diameter (Figure 1(f)) and Θ is the Heaviside function. (iii) *Capillary attraction*: $U_c = -\frac{B}{r^4+1}$ [39] to model quadrupolar capillary attraction. Here, B represents attraction strength, and unity is added to the denominator to prevent divergence at small r [37]. (iv) *Electrostatic (dipolar) repulsion*: $U_d = p/r^3$ [30] describes the long-range dipolar repulsion with strength p .

Table 1: Simulation parameters for different microgel types.

Case (microgel type)	Corona thickness d	Dipolar strength p
I (soft)	0.35	0.2
II (intermediate)	0.22	3.0
III (stiff)	0.05	6.0

To corroborate the effective potential in Eq. (1), we performed MD simulations in LAMMPS [40], using an NVT ensemble and a Langevin thermostat (Supplementary Material, ST3). The following parameters were kept fixed: $\sigma_c = 1.0$, $W_0 = 8.0$, $\lambda_0 = 0.01$, $B = 10.0$, and $S = 19.0$. Consequently, only the parameters d and p were varied to model the microgels used in experiments (see Table 1 where Cases-I, II, III represent microgels of increasing stiffness/elasticity). Since higher contents of the polar crosslinker during synthesis produces microgels with thinner coronas [41] and higher surface charge densities [11], we mapped increasing microgel elasticity to reduced d and increased p . The choice of parameters was based on the particle type, while ensuring that the experimentally observed structures were replicated by tuning a minimum number of parameter values. Since all the self-assembled morphologies could be modeled using this approach, additional contributions of drying-induced capillary and Marangoni flows were not considered. We note here that our model is phenomenological and the simulation parameters were varied based on reports in the available literature [11], [41] rather than direct experimental evidence.

Figure 3(a)–(c) display $U(r)$ versus r for the simulated microgels. Each potential features a primary minimum at a very short interparticle distance, followed by a

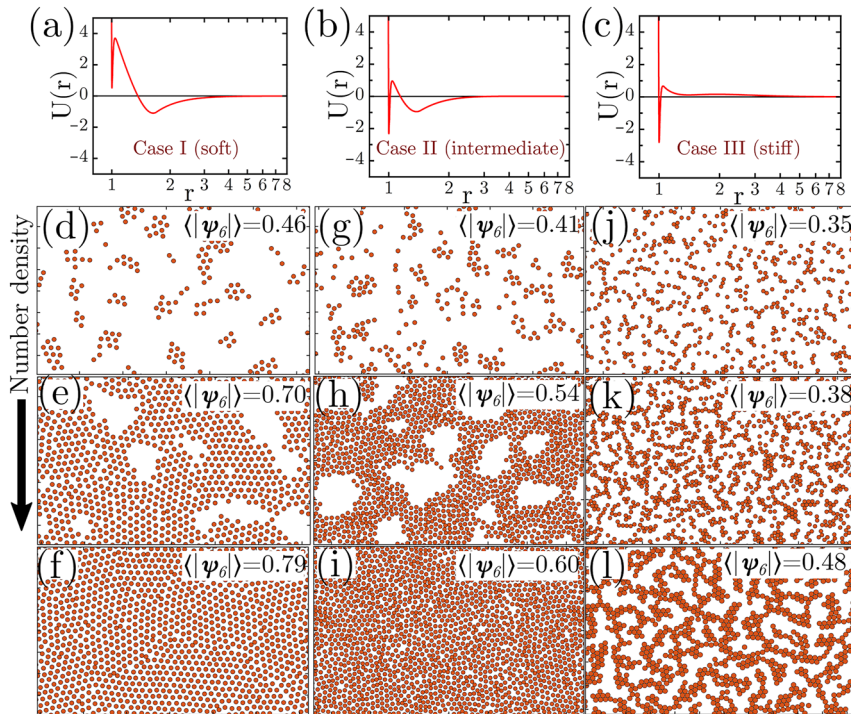


Figure 3: Effective pair potentials, $U(r)$ (in units of $k_B T$), as a function of interparticle separation, r , for (a) case I: soft microgels, (b) case II: microgels of intermediate elasticity and (c) case III: stiff microgels. See Table 1 for the values of corona thickness, d , and dipolar strength, p , used to generate these $U(r)$ curves. Snapshots obtained from MD simulations showing distinct microgel assemblies with increasing particle number density (top to bottom), for (d–f): case I, (g–i): case II, and (j–l): case III.

repulsive barrier and a secondary minimum. As seen from Figure 3(d)–(f) (Case I simulating soft microgels) and Supplementary Material Movies (SV7–SV9), we observe ordered clusters, voids, and non-close-packed hexagonal lattices as the microgel number density increases. While simulating microgels of intermediate stiffness (Case II), we note self-assembled structures that resemble those seen in Case I, however, these morphologies becoming increasingly disordered at the highest density values (Figure 3(g)–(i)). Simulations of stiff microgels (Case III) initially show chain-like structures that percolate and transform to 2D gel-like morphologies (Figure 3(j)–(l), Supplementary Material Movies SV10 and SV11) with increase in density. Our MD simulations therefore capture all the experimentally observed self-assembled morphologies and crossover sequences, thereby providing valuable physical insights. Soft microgels get trapped in the secondary minimum, determined by the competition between capillary and steric interactions, while dipolar interaction remains weak. For stiff microgels in contrast, the primary minimum (due to hydrophobic attraction and core-core hard sphere repulsion) becomes prominent as strong dipolar interactions overwhelm the other components. For the intermediate stiffness case, we have two competing minima that can potentially lead to frustration in a monodisperse assembly. While the morphologies formed by soft microgels are characterized by interparticle

separations that exceed σ_c (Figure 2(a)–(f), and Figure S11 of the Supplementary Material), average interparticle separations in stiff microgel assemblies are $\sim \sigma_c$ (Figure 2(m)–(r), and Figure S11 of the Supplementary Material).

5 Discussion

We computed the pair correlation functions, $g(r)$ (Figure 4(a) and (b)), to compare the structural features of the self-assembled microgel morphologies (at the highest packings) obtained in our experiments and simulations. The experimental curves (Figure 4(a)) reveal a clear crossover from crystalline to disordered packings as microgel elasticity increases. Soft microgels exhibit sharp primary and secondary peaks that are typical of ordered assemblies, while microgels of the intermediate stiffness value show weakened peaks, implying reduced translational order. Stiff microgels display a rapid decay in $g(r)$, consistent with the observed disordered assemblies. The MD simulations reproduce the qualitative trends in $g(r)$ and show a crossover from sharp crystalline peaks (Case I representing soft microgels) to rapidly decaying disordered profiles (Case III corresponding to stiff microgels). Unlike experimental systems, which are typically more polydisperse, the simulated microgel system

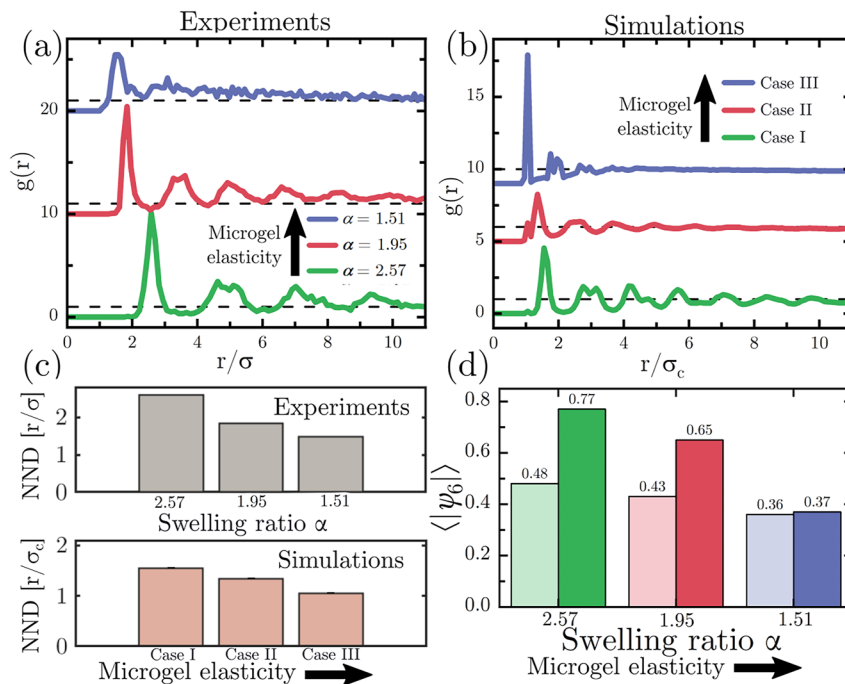


Figure 4: Pair correlation functions, $g(r)$, obtained from (a) microscopy images and (b) MD simulations. (c) Nearest neighbor distance (NND) as a function of microgel elasticity from experiments and simulations, respectively. Highest ϕ regions (Figure 2(f), (l), (r) and Figure 3(f), (i), (l)) were used to calculate $g(r)$ and NND. (d) Evolution of $\langle |\psi_6| \rangle$ for different microgel elasticities computed for the highest ϕ regions in Figure 2. Figure 2(e), (f), (k), (l), (q), and (r) were analyzed to calculate $\langle |\psi_6| \rangle$ at early and late times (light and dark color bars respectively).

is perfectly monodisperse. Because the shape of the primary minimum (Figure 3(c)) dictates the profile of the first peak, the sharper peaks observed in the simulations directly result from the sharp primary minimum of the model potential. Further inspection of Figure 4(a) and (b) reveals a decrease in nearest neighbor distances as particle elasticity increases in both experiments and simulations (Figure 4(c)). These features substantiate our earlier observations (soft microgels assemble in loose, ordered structures, while stiff particles form close-packed, disordered aggregates) and provide robust validation for our microscopic model. The global ordering of microgels of different elasticities at the highest ϕ values (Figure 2(e), (f), (k), (l), (q), and (r)) was quantified by plotting $\langle |\psi_6| \rangle$ in Figure 4(d) at early and late times (denoted respectively by light and dark color bars). While soft microgel particles exhibit an increase in structural order over time, stiff microgel assemblies, kinetically trapped in chains and percolating networks, do not show any time-evolution. Even though we may intuitively expect that stiffer microgels exhibit stronger steric repulsion, our results show that the steric interaction range (controlled by corona thickness d) dominates over steric strength (S) in determining the observed interfacial morphologies. Remarkably, a minimal two-parameter variation in the effective interparticle potential (d , p) captures the full spectrum of microgel self-assembly, from ordered lattices to disordered gels, in the highly nonequilibrium, time-evolving environment studied here.

6 Conclusions

Our work shows that the rich variety of microgel morphologies observed at the air-water interface of a drying droplet is critically sensitive to particle elasticity (determined by steric and dipolar interactions). MD simulations, implemented with a minimal effective double-well potential, successfully capture all experimentally observed morphologies. We identify particle elasticity as a control parameter that can fine-tune effective interparticle interactions and the nonequilibrium self-assembly of deformable colloids at a fluid interface. Our findings reinforce a growing consensus [42] that the mechanical properties of individual constituents (e.g., elasticity) govern the emergent structures and phase behavior of soft and biological matter assemblies. In future work, it will be interesting to explore the roles of the two competing length scales observed in the intermediate-stiffness regime. Such studies may offer a unique experimental platform for studying emergent mechanical responses and glass physics in monodisperse soft colloidal materials. Since our

simulation parameters were not experimentally measured, direct determination of interparticle potentials using optical tweezers [43] or machine learning [44] would provide quantitative validation of interaction ranges and strengths.

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