

Deformation regimes in soft materials under large amplitude oscillatory shearRishav Agrawal ^{*}*School of Engineering, University of Liverpool, United Kingdom*Patrick T. Spicer [†]*School of Chemical Engineering, University of New South Wales, Sydney, New South Wales 2052, Australia*Esther García-Tuñón [†]*School of Engineering and Materials Innovation Factory, University of Liverpool, United Kingdom*

(Received 21 November 2025; accepted 14 May 2026; published 11 June 2026)

Three-dimensional printing of advanced functional materials via direct ink writing requires the design and processing of soft solids or yield stress materials with high solids concentrations. The deformation, flow, and microstructure evolution of such complex soft materials are central in additive manufacturing extrusion technologies. In this study, we investigate both bulk properties and microscopic dynamics using rheo-microscopy, particle tracking, and birefringence mapping. This work presents an integrated framework that maps bulk rheology to local spatiotemporal heterogeneities during yielding of soft materials by combining oscillatory rheology experiments with simultaneous *in situ* imaging. Using a Pluronic F127 hard gel matrix with increasing concentrations of silica fillers as a model for direct ink writing formulations, we quantify the evolution of deformation pathways under large-amplitude oscillatory shear tests with increasing concentration (Φ). Mapping local events onto bulk rheological metrics unveils a consistent trend from a matrix-dominated to a filler-dominated deformation regime. At low concentrations ($\Phi \lesssim 0.31$), the matrix dominates the yielding mechanism, leading to an affine-banding-fracture pathway. However, at high concentrations ($\Phi > 0.31$), the filler-dominated system undergoes multiple local events under small deformations, from which cracks propagate and fracture the material into yielded and solid like regions. This work reconciles bulk and local observations in a flow regime map for particle suspensions in a gel matrix, highlighting when a soft material exhibits the desired affine flow and when it undergoes catastrophic failure. The results demonstrate that the deformation of highly concentrated composites is complex, where heterogeneous yielding and fracture coexist and negatively impact printability and shape fidelity. The experimental evidence presented in this work provides a timely avenue toward understanding the heterogeneous deformation and flow of complex soft materials, which play a critical role in a wide breadth of applications beyond 3D printing.

DOI: [10.1103/d86k-mgq6](https://doi.org/10.1103/d86k-mgq6)**I. INTRODUCTION**

Soft materials such as paste like concentrated suspensions and yield stress materials are central to many applications, including direct ink writing (DIW) [1], foods, pharmaceutical industries [2], drilling fluids [3], and natural debris flows [4]. Understanding the deformation and flow of soft materials remains an open research question in condensed matter physics [5,6]. The solid like to liquid like or yielding transition is ubiquitous across many disciplines from materials science and engineering, soft matter physics, and non-Newtonian fluid mechanics to advanced materials manufacturing. Bulk rheological techniques are frequently used to quantify the yielding

transition. For example, large amplitude oscillatory shear (LAOS) experiments provide the material's stress response to a sinusoidal strain input with increasing amplitude at fixed oscillation frequency [7,8]. Many soft materials, such as colloidal glasses, emulsions, polymers, and surfactants display similar bulk stress responses with increasing strain amplitudes, e.g., crossover of storage G' and loss G'' moduli, and G'' overshoot. These common observations have motivated researchers to pursue a unified description of the yielding transition. The early works of Bingham [9] in the 1920s, and more recent advanced constitutive models (Saramito's elasto-visco-plastic model [10] and Kamani-Donley-Rogers (KDR) models [11,12]) aim to capture the yielding transition under continuum approximations. The modified KDR model introduces a britility factor (B_t parameter) [12] to predict a spectrum of behaviors, ranging from ductile to brittle yielding. However, models based on homogeneous flow assumptions cannot capture the rich microstructural heterogeneous evolution these soft materials undergo during deformation.

The compositions and microstructure of soft materials are extremely diverse, creating barriers to the development of

^{*}Contact author: ragrawal@liverpool.ac.uk[†]Contact author: egarcia@liverpool.ac.uk

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

universal physical frameworks [11,13]. This diversity is clearly exemplified in the DIW context [14–19]. For example, hydrogels used in bioprinting applications include alginate, gelatin derivatives, collagen, or hyaluronic acid [20,21]. Advanced materials formulations often require high solids loading and different additives to meet rheological and functional requirements [16,17,19,22]. F127 is a poly-ethylene-oxide and poly-propylene-oxide (PEO-PPO-PEO) triblock copolymer which assembles into micelles and further orders into structured mesoscopic phases in solution [23]. Each micelle, with a core dominated by PPO and a corona dominated by hydrated PEO, is considered a hard sphere [23,24]. The F127 lyotropic liquid crystalline (LLC) phases are highly dependent on solvent, concentration, temperature, and shear [24–29] and are widely used in drug delivery, tissue engineering, and in DIW [22,30]. F127's cubic phase, I_1 (25 wt %, and $\sim 18^\circ\text{C}$) can become easily disrupted leading to heterogeneous domains within the material [29]. The temperature-dependent F127 micellar phases have been described as soft and hard gels from a rheological perspective [31]. The latter are commonly used as carriers or matrices to suspend high concentrations of functional materials for DIW applications [18,22,30,32,33]. The combination of a matrix that is a complex fluid itself and high solids loading adds up to form hierarchical structures where deformation and flow are governed by interactions at multiple length scales (Sec. II A).

Direct imaging in rheological experiments has revealed that yielding of these soft materials varies significantly with the underlying microstructure and is not spatially homogeneous [29,34–37]. Instead, yielding takes place via localized rearrangements, transient shear bands, and fracture like events that bulk rheology cannot distinguish. Preparation history can also play a major role in the yielding process; for example, increased annealing drives the material's structural evolution from progressive (ductile like) to abrupt, banding-dominated (brittle like) yielding [38,39]. These observations indicate that the microscale structure dominates the yielding pathways in soft materials, highlighting the limitations of bulk rheological techniques that inherently assume a homogeneous flow field within the sample [40,41]. LAOS analysis frameworks, such as Fourier transform (FT) rheology and the sequence of physical processes (SPP), provide quantitative measures of nonlinearity [3,8,42]. However, they cannot directly resolve local events such as shear banding, wall slip, or fracture [35,36]. Rheo-microscopy of Pluronic F127 at concentrations that form micellar cubic phases [25] and hard gels [29] evidences that the flow becomes heterogeneous under oscillatory deformation, leading to a fracture like event at the strain amplitudes that bulk rheology would label flowing or liquid like.

Noncontinuum frameworks such as soft glassy rheology (SGR) and mode-coupling theory (MCT) capture the microscopic dynamics in soft materials [43–46]. Despite their success, SGR and MCT struggle to quantitatively predict bulk rheology and LAOS responses because they lack spatial resolution, realistic ageing dynamics, and a unified description across glass and jamming transitions [47]. Elastoplastic models describe soft materials as elastically deforming mesoscopic blocks that yield plastically once the local stress exceeds a threshold. These models provide insights into the

strain localization as well as yielding transition and the statistics of avalanches of plastic events [48–52]. Recent particle-resolved simulations of dense frictionless suspensions and granular materials [53,54] have introduced alternative scaling laws and identified nonmonotonic constitutive behavior. These significant advances move beyond the assumption of homogeneous shear and provide a framework to understand shear banding. However, these studies have so far been focused on Newtonian carrier fluids and do not capture the non-Newtonian rheology of suspending matrices such as the complex liquid crystalline matrix in this work. Here, we provide an experimental study that can serve as a source for the validation or further development of theoretical studies from different perspectives.

From an applied experimental perspective, the yielding transition plays a significant role in three-dimensional (3D) printing via DIW, which relies on the extrusion of soft materials. Ideal DIW inks at rest behave like an elastic solid, maintaining the desired shape, yet they flow smoothly through narrow nozzles under applied stress [1,55–58]. Increasing the content of active functional materials such as graphene oxide [14], alumina [32], and graphite [30] increases small-amplitude storage modulus (G'_{LVR}) [14], flow stress (σ_f), and the material strength [18]. However, increasing solid loading compromises the flowability and recoverability and, as a consequence, the shape fidelity of the final printed structures [30].

Entwined questions arise: How do these active materials modify the yielding transition? What causes the loss of print fidelity with increasing content of active material? Linking bulk metrics with microstructural information is necessary to uncover the underlying local phenomena leading to loss of printing accuracy [29,30]. Here we answer these questions using a model system: a Pluronic F127 liquid crystal as a background hard gel matrix [29,31], mixed with increasing concentrations of monodisperse silica (SiO_2) fillers with volume fraction Φ . This provides a single, controlled parameter that switches the composite's yielding pathways. Comparing LAOS bulk data using FT rheology and the SPP framework (Fig. 1) against direct imaging results enables distinguishing affine flow from heterogeneous banding, crack formation, and brittle fracture. Affine flow means the flow profile matches the imposed shearing deformation, while banding means that the material is deformed unevenly, and the flow deviates from the expected linear profile. Polarized microscopy observations enable us to map structural heterogeneity and transitions due to the crystallization and disruption of the F127's micellar phase.

II. METHODS

A. Sample preparation

BASF Pluronic F127 (Sigma-Aldrich, BioReagent) was used as the liquid crystalline matrix. Stock solutions were prepared by dispersing F127 powder into deionized water at room temperature ($\approx 22^\circ\text{C}$) using a planetary mixer (THIARE250, Thinky). The mixer was operated in alternating mixing (2000 rpm, 2 min) and defoaming (2000 rpm, 2 min) cycles, with intermittent cooling in an ice bath to prevent overheating. Samples were stored at 3°C for at least 24 h before

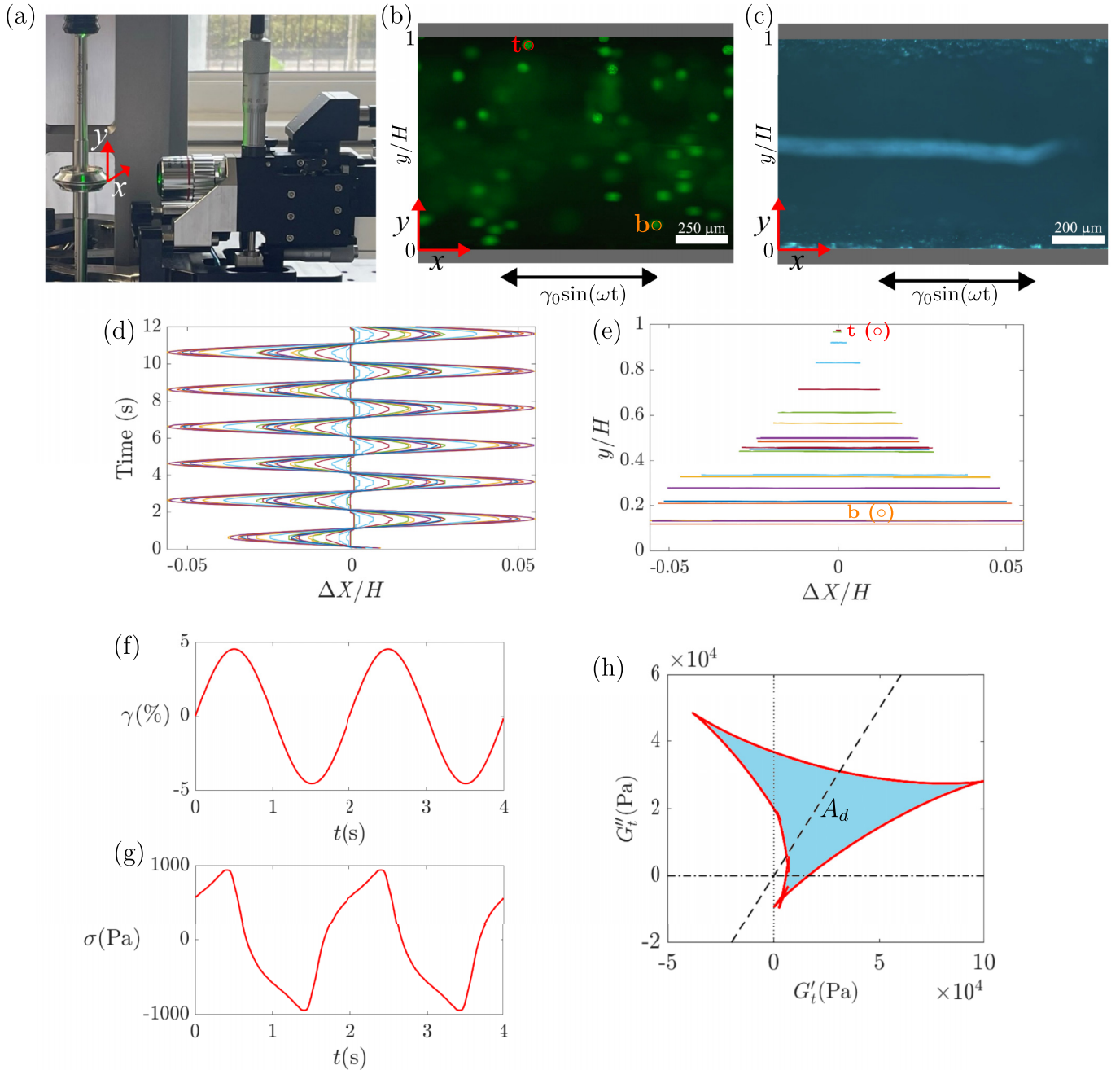


FIG. 1. Overview of the experimental setup and data analysis. (a) Twin-drive AP MCR702e Rheometer with a sandblasted parallel-plate geometry. The microscope is located on the right side. The x - y coordinates are included as a point of reference. (b), (c) Microscopy images taken at the edge of the gap. The shear direction is indicated by the red horizontal arrow (x). The gap (H) used was 1 mm for fluorescence microscopy (b) and 0.8 mm for polarized microscopy (c). The green spheres in (b) represent fluorescent tracers used for particle tracking measurements. A tracer at the top is marked with a t ($\circ$) and another at the bottom with a b ($\circ$) as a guide to visualize their displacements in graph (e). (d), (e) Particle tracking results are shown as projections of the time-dependent tracer displacement trajectories for $\Phi = 0.05$, where $\Delta X = X - \langle X \rangle$, and $\langle X \rangle$ is the average displacement. (f), (g) Representative LAOS strain and stress time series ($\Phi = 0.05$) analyzed using FT rheology and the SPP framework. (h) Representative SPP Cole-Cole plot ($\Phi = 0.05$) used for the calculation of the deltoid area (A_d) within the $[G'_t, G''_t]$ trajectory (Sec. II B 1).

use to ensure degassing as well as identical thermal history, following protocols previously established [29,30]. Spherical silica particles (8–12 μm , uncoated glass, Cospheric) were added to the F127 stock solution at concentrations of 10, 30, 50, 60, and 70 wt%. Particles were added in multiple steps to the F127 formulation base in its soft gel state [31]

below the micellar transition temperature, with mixing and cooling steps in between SiO_2 powder additions. It is expected that the silica particles will primarily interact with the F127 matrix through hydrogen bonding between the silanol ($-\text{SiOH}$) groups on silica particles and the hydrated PEO blocks on the corona micelles [23,59,60]. For particle track-

ing (PT) experiments, fluorescent polyethylene microspheres (53–63 μm , Cospheric) were added at ~ 0.15 wt % to the formulations before the final mixing and cooling step. The filler volume fraction, Φ , was calculated from the mass fraction of silica fillers, w , and the densities of the filler (ρ_f) and matrix (ρ_m) according to

$$\Phi = \frac{w/\rho_f}{w/\rho_f + (1-w)/\rho_m}, \quad (1)$$

where $\rho_f = 2.2 \text{ g cm}^{-3}$ for silica and $\rho_m = 1.0 \text{ g cm}^{-3}$ for the F127-water matrix at room temperature. Using Eq. (1), the 10, 30, 50, 60, and 70 wt % formulations correspond to $\Phi = 0.05$, $\Phi = 0.16$, $\Phi = 0.31$, $\Phi = 0.41$, and $\Phi = 0.51$, respectively. Although $\Phi = 0.51$ is below the random close packing limit of dry hard spheres (~ 0.64), contact-dominated stress transmission occurs at substantially lower volume fractions in concentrated suspensions and filled soft solids [61,62], making this concentration significant in the present system. Tracer particles (< 0.15 wt %) were neglected in the calculation. The SiO_2 -F127-water mixture can be regarded as a composite lyotropic colloidal suspension. The F127 matrix with a concentration of 25 wt % is a hard gel [31] and a transparent, paste like material, owing to the arrangement of colloidal hard spheres in a cubic arrangement [23], which traps the silica fillers [63]. Given the density difference, the particles are subjected to gravitational forces [$F_g = \frac{4}{3}\pi a^3(\rho_f - \rho_m)g$, where a is the particle radius, ρ_f is the filler density, and ρ_m is the matrix density, $F_g \sim 0.1$ Pa]. However, the yield stress (between ~ 250 and 1000 Pa, depending on Φ) of the matrix and composites are orders of magnitude higher, preventing sedimentation in the hard gel state. The network formation is governed by the micellar structure and the hydrogen bonds between the silanol and the PEO corona. The strength of this network is evidenced by the large storage modulus in the linear viscoelastic region (> 10 kPa for the matrix, and > 100 kPa for the highest Φ) and the critical strains observed in LAOS (Sec. III A).

B. Rheo-microscopy measurements

Rheo-microscopy experiments were performed on an Anton Paar MCR 702 rheometer in a twin-drive mode with air-bearing-supported motors [Figs. 1(a)–1(c)] [29]. Sand-blasted stainless steel parallel plates (diameter $2R = 40$ mm) were used with a gap of $H = 1.0$ mm for particle tracking experiments and $H = 0.8$ mm for birefringence measurements. LAOS tests were carried out using strain amplitude sweeps ($\gamma_0 = 0.01$ – 100%) at a fixed frequency, $f = 0.5$ Hz. Sinusoidal strain waveforms were set via the custom waveform generator option instead of the built-in LAOS template to ensure precise control of the oscillation cycles. Stress responses were recorded for nine oscillation cycles at each strain amplitude γ_0 , and the last two full cycles were used for analysis [Figs. 1(f) and 1(g)]. Each concentration was characterized in quadruplicate to ensure the reproducibility of the results.

1. LAOS analysis of raw waveforms

LAOS data are analyzed using FT rheology [64], and the SPP framework [42] (Fig. 1). FT rheology is used to calculate the first harmonics of the Fourier-transformed stress response,

providing the G' and G'' values, also known as storage and loss moduli, respectively. The nonlinear onset strain (γ_{nl}) is defined from $I_{3/1} > 0.01$ [30], where $I_{3/1}$ is the intensity ratio of the third to the first harmonics of the stress response. The dissipation ratio (ϕ) [64] calculated from the FT-analysis outputs [18] is used to compare the energy dissipated in a single oscillation cycle [area enclosed in an elastic Lissajous-Bowditch (LB) curve, E_d] to the energy that would be dissipated by a theoretical perfectly plastic material ($E_d)_{pp}$, given by $\phi = E_d/(E_d)_{pp}$ [64]. The SPP framework provides instantaneous information at each point in the stress-strain-strain rate trajectories through the transient elastic ($G'_t = \partial\sigma/\partial\gamma$) and viscous ($G''_t = \omega\partial\sigma/\partial\dot{\gamma}$) moduli [42,65,66]. The intracycle responses are generally analyzed through the $[G'_t, G''_t]$ trajectories in a Cole-Cole plot during each oscillation cycle [Fig. 1(h)]. The position, shape evolution, and area (A_d) of the deltoids in the Cole-Cole plots can be associated with different physical processes [42,65,66]. The area of each deltoid A_d is a measure of energy dissipation associated with microstructural arrangements taking place in a given cycle [42,65,66].

2. Image acquisition

In situ imaging via fluorescence microscopy and polarized-light microscopy was performed during LAOS tests using a side-mounted microscope [Fig. 1(a)]. Fluorescence microscopy was performed with a $5\times$ objective and a Sugar-CUBE LED illuminator, recorded at 15.19 fps with a ~ 1 mm field of view [Fig. 1(b)]. Polarized-light microscopy was performed by inserting a linear polarizer into the optical path instead of the band-pass filter to study birefringence during oscillatory shear [Fig. 1(c)]. The acquired images are synchronized with bulk rheological data via custom MATLAB scripts.

C. Particle tracking and birefringence analysis

The methodology is illustrated in a schematic workflow that summarizes experimental and analysis methods (Fig. 1). Particle tracking is carried out using the TrackMate plugin in Fiji [67,68]. Between 20 and 30 tracers are tracked per experiment. Transient displacement profiles are extracted over the entire nine cycles for each strain amplitude [Figs. 1(d) and 1(e)]. Additional LAOS experiments were performed to analyze birefringence in reflection using a polarized rheo-microscopy setup [29] (at $\Phi = 0.05$, 0.16 , and 0.51). A linear polarizer and analyzer were set at intermediate angles to enhance sensitivity to weak elliptical polarized light. The images are synchronized with strain input and time, and analysed using custom MATLAB scripts. The mean intensity profiles for each row in each image are extracted and normalized with respect to the maximum intensity in each dataset. The normalized intensity is plotted against a dimensionless time (Fig. S4 in the Supplemental Material (SM) [69]). These intensity maps enable the quantification of structural transitions during the LAOS test, distinguishing between dark (isotropic) and birefringent (anisotropic) regions as the crystallization of the cubic structure takes place [29].

D. Direct ink writing

DIW of the SiO_2 -F127- H_2O composite formulations ($\Phi = 0.05$ and 0.51) was performed using a custom-built robocaster equipped with motorized syringe plungers [14,70]. The Aerotech A3200 motion composer software controls the X-Y-Z motion of the Gantry robot using G-code. The F127-water matrix and SiO_2 -F127-water composites were loaded into 5 mL syringes (Nordson, EFD) using a spatula, ensuring that no air bubbles were trapped. Each syringe was fitted with a Luer lock stainless steel nozzle tip with an inner diameter of 0.51 mm. Stepper motors were used to control the plunger displacement, to feed the material to the nozzle tip before printing, and maintain a constant flow during printing.

A lattice grid structure was designed in the RoboCad 5.0 software and converted to G-code for printing (Fig. S1 in the SM [69]). The generated G-code was executed by the Aerotech motion controller to deposit filaments directly onto PTFE substrates. A dummy lead-in line (~ 25 mm) was printed prior to each structure to stabilize the flow. Printing was carried out at room temperature ($\approx 22^\circ\text{C}$) using the same nozzle and motion settings for both $\Phi = 0.05$ and 0.51 to enable direct comparison. No postprinting drying study was carried out, as the focus here was limited to assessing structure stability and shape fidelity immediately after extrusion.

III. RESULTS

A. Trends of bulk metrics with increasing Φ

Bulk rheological metrics do not directly reveal quantitative measures of spatial heterogeneity. However, they show a gradual and systematic evolution with filler concentration caused by a shift in microstructure and microscale dynamics (Fig. 2). The strength of the composite F127- SiO_2 model systems increases with solids loading (Φ), as evidenced by the trends in stress amplitude, storage modulus, and loss modulus [σ_0 , G' , and G'' in Figs. 2(a)–2(c), respectively]. G'' increases almost two orders of magnitude, while losing its characteristic overshoot with increasing solids loading. This shift is consistent with F127-Graphite trends previously reported [30]. Other bulk measures also match the observed LAOS fingerprints for F127-Graphite formulations [30]: the linear regime shrinks with an early onset of nonlinearity [γ_{nl} , $I_{3/1}$, Fig. 2(d)] and the energy dissipation increases with increasing solids loading. The latter is quantified by the dissipation ratio [ϕ_{LVR} , Fig. 2(e)] and the area of the deltoids in the SPP Cole-Cole plots [$A_{d,\text{max}}$, Fig. 2(f)]. The bulk LAOS metrics (Figs. 2 and 3), the Lissajous-Bowditch plots, and the intra and intercycle trajectories in the Cole-Cole plots for the model system (Fig. S2 in the SM [69]) show consistent trends linked to microscopic events.

Here we investigate the underlying microscopic dynamics and yielding pathways responsible for the observed trends with increasing filler concentration (Φ , Sec. III B). The combined bulk and direct imaging results reveal two different mechanical regimes as the solid loading increases:

(1) A matrix-dominated regime at low and moderate filler concentration ($\Phi \lesssim 0.31$) where the F127 matrix governs the response (Sec. III B 1).

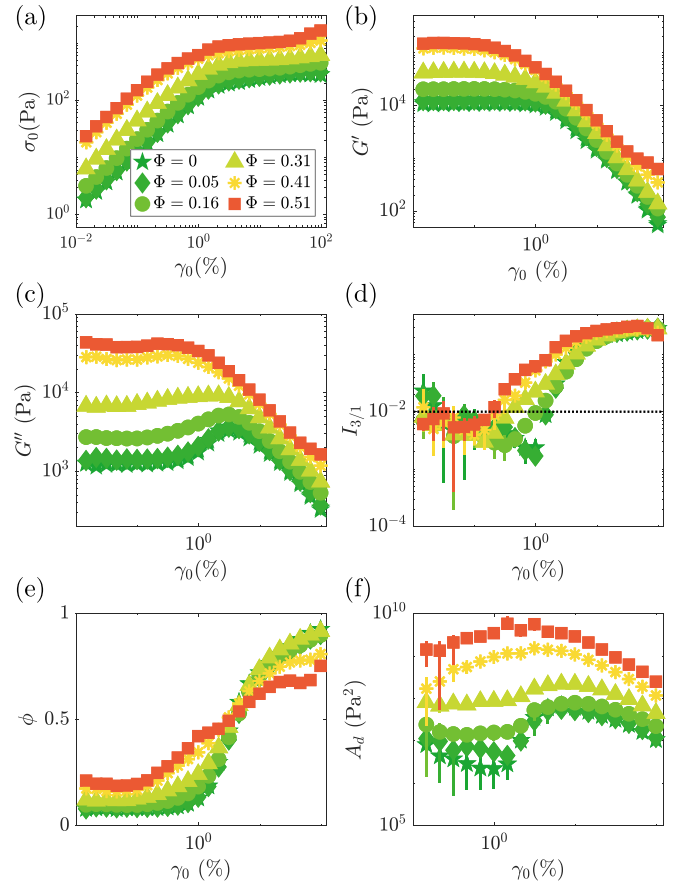


FIG. 2. Bulk rheological results for the Pluronic F127 hard gel liquid crystalline matrices (25 wt % in DI water) containing SiO_2 powders. The graphs show the large amplitude oscillatory shear (LAOS) responses as a function of strain amplitude γ_0 . (a) Maximum stress extracted from each cycle, (b) storage modulus G' , (c) loss modulus G'' , (d) intensity ratio of the third to the first harmonic of the stress output $I_{3/1}$, (e) dissipation ratio ϕ , and (f) the area of the SPP deltoids A_d obtained from the Cole-Cole plots, which shows the evolution of energy dissipation with strain amplitude due to microstructural rearrangements. The green to red color scheme of the symbols represents the evolution of the regime from matrix-dominated to filler-dominated quantified based on particle tracking results (Sec. III B).

(2) A filler-dominated regime at high solids loading ($\Phi > 0.31$) where the filler network controls the response, leading to early onset of nonlinearities and increased energy dissipation due to plastic events (Fig. 2, Sec. III B 2).

Bulk rheological metrics display a prominent G'' overshoot [Figs. 2(c) and 3(a)] with dissipation ratio values shifting from $\phi_{\text{LVR}} \approx 0.1$ to $\phi_{G'' > G'} \rightarrow 1$ (Figs. 2(e) and 3(c)) under increasing deformations for the matrix-dominated regime (at $\Phi \lesssim 0.31$). The G'' peak is associated with type-III behavior [71], a characteristic feature of many yield stress materials. This overshoot has been explained as a transition from predominantly recoverable elastic to unrecoverable deformations [72,73]. LB curves (Fig. S2 in the SM [69]) approaching a rectangular shape at large strain amplitudes have been associated with perfect plastic behavior [3], corresponding to $\phi = 1$ (Sec. II B 1). Spatiotemporal microscopy enables us to

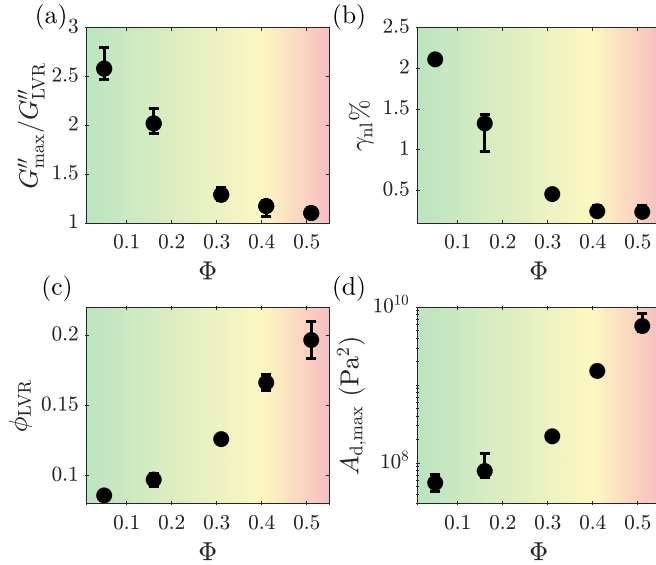


FIG. 3. Rheological metrics as a function of filler volume fraction (Φ): (a) normalized loss-modulus maximum $G''_{\max}/G''_{\text{LVR}}$, (b) strain onset for nonlinearity γ_{nl} , (c) small-amplitude dissipation ratio ϕ_{LVR} calculated at nominal strain value, $\gamma_0 = 0.1\%$, ϕ_{LVR} provides a measure of energy dissipation in the linear solid like regime, and (d) maximum deltoid area $A_{d,\max}$. The green to red color scheme of the background represents the evolution of the regime from matrix-dominated to filler-dominated quantified based on particle tracking results (Sec. III B).

connect these characteristic features with flow instabilities and structural transitions, and to quantify banding coinciding with the onset of nonlinearity and G'' increase, and ϕ and A_d trends with fracture (Sec. III B 1).

In the filler-dominated regime at high Φ , the G'' overshoot is suppressed [Fig. 3(a)], evidencing a change of underlying

energy transitions and shift to a type-I behavior associated with strain thinning or structural breakdown [71]. Energy dissipation metrics increase with solids loading at small deformations with $\phi_{\text{LVR}} \approx 0.24$ [Fig. 3(c)] and A_d increasing up to two orders of magnitude [Fig. 3(d)]. Direct visualization evidences local events and crack initiation at strain amplitudes as small as $\gamma_{\text{fr}} \lesssim 0.67\%$ [Fig. 4(a)] that explain the observed ϕ and A_d trends (Sec. III B 2).

These findings enable the reconciliation of bulk and microscopic flow by establishing quantitative links between affine deformation, banding, and fracture, with specific bulk metrics that evidence concentration-dependent deformation pathways (Sec. III 3).

B. Microscopic heterogeneous flows

PT and birefringence mapping enable the quantification of heterogeneous flows and structural transitions [29]. PT results demonstrate the shift in yielding mechanisms with increasing SiO_2 filler concentration, Φ . The maximum and transient displacement profiles from PT provide a criterion to distinguish the three deformation regimes observed in this study: affine, banding, and fracture (Figs. 4 and 5). Each of these regimes is highlighted in green, yellow, and red, respectively (Figs. 3, 5, and 7). The displacement profiles across the gap remain linear in the affine regime, quantified through R^2 values for a linear fit, with a threshold of $R^2 > 0.995$. The displacement profiles become curved in the banded regime. However, they remain continuous, indicating heterogeneous flow without any abrupt discontinuities in the flow. In the fracture regime, the profiles display intense discontinuities with portions of the profile collapsing onto the same displacement value, i.e., either remaining nearly static (zero displacement) or moving as a rigid solid at the maximum displacement. Flat or step-like regions indicate the coexistence of yielded and

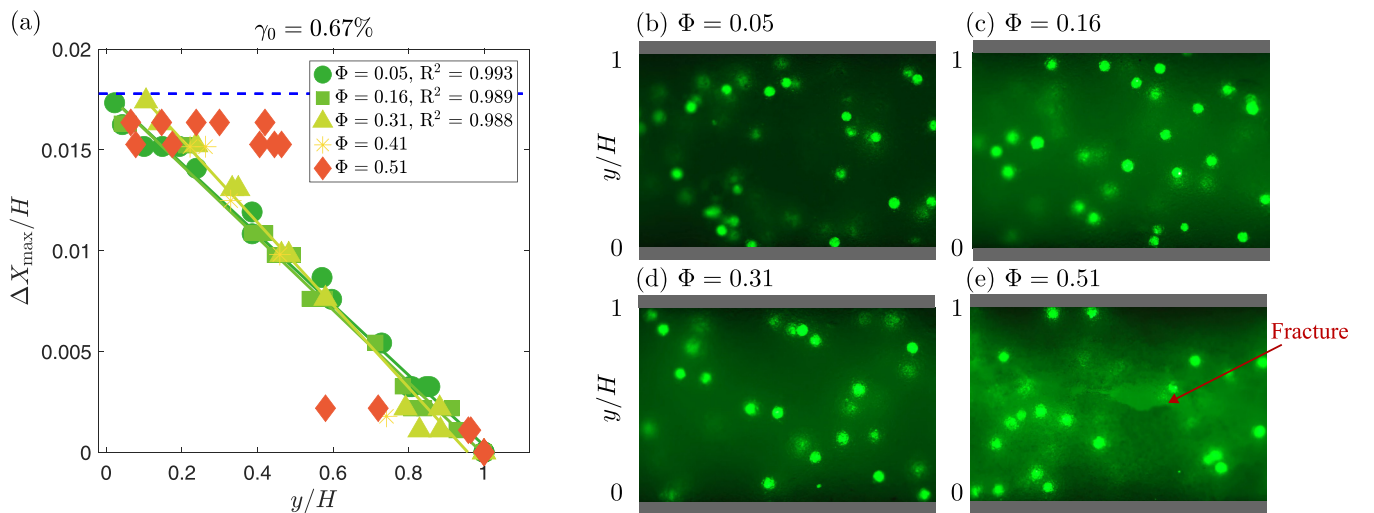


FIG. 4. Deformation profiles and local microstructural evolution in F127 gels containing SiO_2 fillers. (a) Maximum displacement profiles ($\Delta X_{\max}/H$) across the gap at strain amplitude $\gamma_0 = 0.67\%$ for different filler volume fractions $\Phi = 0.05, 0.16, 0.31$, and 0.51 . Linear fits (R^2) are used to quantify the deviation from affine motion. (b)–(e) Fluorescence microscopy frames acquired at $\gamma_0 = 0.67\%$, showing homogeneous tracer displacements up to $\Phi = 0.16$ and a fracture for $\Phi = 0.51$. The green spheres in (b)–(e) are the tracer particles used for fluorescence microscopy.

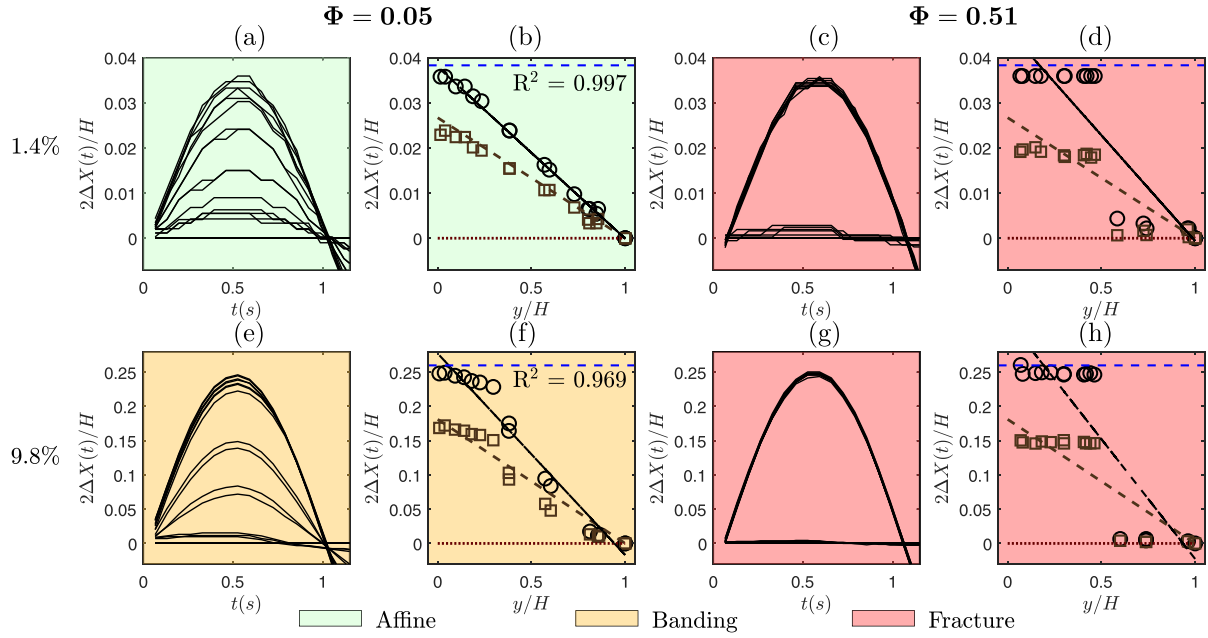


FIG. 5. Transient deformation of F127-silica gels at two strain amplitudes [top: $\gamma_0 = 1.4\%$ (a)–(d), bottom: $\gamma_0 = 9.8\%$ (e)–(h)]. Odd columns (a), (c), (e), (g) show time-resolved displacement profiles $\Delta X(t)/H$ across the gap over a half-cycle. Even columns (b), (d), (f), (h) show the same data sampled at two phases of the cycle ($\square$ at $t = T/8$, and $\circ$ at $t = T/4$, which corresponds to maximum displacement), overlaid with least-squares trends. R^2 values correspond to the maximum displacement profiles. The dashed and solid lines correspond to the theoretical values calculated from the strain values at $t = T/8$ and $T/4$ phases, respectively (where T is the time-period of oscillation). The shaded backgrounds denote each of the three regimes: affine in green, banding in yellow, and fracture in red.

unyielded zones within the material. Such discontinuities are associated with fractures within the sample. This framework, based on displacement profiles, provides a consistent and reproducible way to separate affine deformation, banding, and fracture to reconcile bulk trends and microscopic dynamics (Sec. III C). The tracer's displacement profiles clearly distinguish concentration-dependent deformation pathways from near-affine deformation at low solids loading ($\Phi \leq 0.31$) to discontinuities at high concentrations ($\Phi = 0.51$) due to fracture and slip [at $\gamma_0 = 0.67\%$, Fig. 4(a)].

1. Matrix-dominated regime

Rheo-microscopy results for the matrix-dominated SiO₂-F127-water mixtures show that $I_{3/1}$ trends are reliable indicators of the onset of yielding, which coincides with the transition from affine deformation to banding at γ_{nl} [Figs. 2(d) and 3(d)], while the near-rectangular LB shapes at large strain amplitudes correspond to fracture in the so-called liquid like regime (when $\gamma_0 \uparrow$, $\phi_{G'' > G'} \rightarrow 1$, Fig. S2 in the SM [69]). The coexistence of unyielded and yielded regions within the same material [29] does not represent the idealized perfect plastic like response [3]. The KDR model [11] can predict the LAOS response of the F127 matrix without SiO₂ fillers [74], but it cannot yet capture microscopic dynamics (Figs. 4 and 5). In principle, bulk rheology cannot distinguish idealized yielding from heterogeneous flow or fracture since the measurements are made on the assumption of ideal flow conditions. Here, PT shows that there are specific bulk rheological features that can be connected with fracture (Sec. III C).

Linear displacement profiles across the gap demonstrate that affine deformation takes place at small strain amplitudes [e.g., at $\Phi = 0.05$, Fig. 4(a)]. The transient displacement signals vary smoothly for each tracer [Fig. 5(a)] and scale linearly with y/H , with a high goodness-of-fit linear profile [$R^2 \approx 0.997$, Fig. 5(b)]. Fluorescence microscopy results do not show evidence of discontinuities or flow instabilities, in agreement with bulk measurements [Figs. 4(b)–4(d)]. When the strain amplitude approaches γ_{nl} , the displacement profiles deviate from affine deformation, which evidences the onset of spatial heterogeneity ($R^2 \downarrow$, Fig. S3 in the SM [69]). As γ_0 increases above the nonlinear onset, the deviations intensify into banding. The tracers' transient signals split into four groups [Fig. 5(e)], and the normalized displacement profiles show a reduced goodness-of-fit R^2 [Fig. 5(f)]. At large amplitudes beyond the moduli crossover ($G' = G''$), these bands break up into yielded and unyielded regions. The fracture splits the material into a region that undergoes rigid solid motion in sync with the moving geometry, while the other remains almost static, attached to the stationary plate (supporting videos in the SM [69]). A dilation like effect seems to originate from the center of the gap with increasing strain amplitudes (fluorescence videos for PT in the SM [69]). This behavior could be consistent with normal-stress driven out-of-plane displacement during yielding [75,76] and microstructural reorganization of the F127 matrix. With increasing solids loading ($\Phi \uparrow$), this effect becomes more pronounced due to enhanced crowding of filler particles (fluorescence videos for PT in the SM [69]). Birefringence mapping of the matrix-dominated F127-SiO₂ mixtures evidences consistent

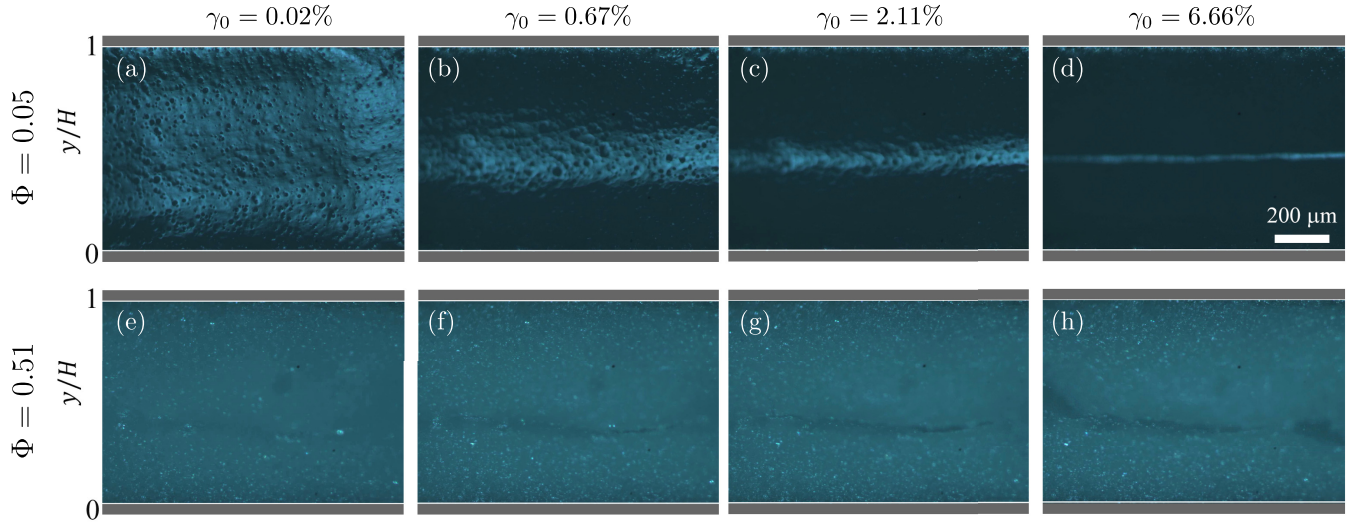


FIG. 6. Polarized microscopy images for matrix ($\Phi = 0.05$) and filler-dominated ($\Phi = 0.51$) systems at strain amplitudes $\gamma_0 = 0.02\%$ (a), (e), 0.67% (b), (f), 2.11% (c), (g), and 6.66% (d), (h). For $\Phi = 0.05$, the images show the progression of dark regions [at $\gamma_0 = 0.67\%$ (b) and 2.11% (c)] due to the crystallization of the F127 cubic structure from the measuring plates, and the displacement of a birefringent band toward the middle of the gap (Fig. S4 in the SM [69]). At $\gamma_0 = 6.66\%$ (d), most of the gap becomes dark. A faint central anisotropic band remains where the fracture occurs. (e)–(h) Above $\Phi \approx 0.31$, it is not possible to map the crystallization of the cubic structure, likely due to multiple light scattering masking the form birefringence. (e) Local events take place at $\gamma_0 = 0.02\%$ for $\Phi = 0.51$, with fillers rotating and sliding against neighboring particles. (f), (g) Cracks initiate from these regions and propagate as strain amplitude increases, leading to complex fractures (h).

structural transitions due to the crystallization of the cubic phase from the measuring plates (Figs. 6(a)–6(d) and S4 in the SM [69]), and the displacement of an anisotropic birefringent structure [77,78] toward the center of the gap [29]. This surface-driven crystallization concentrates the birefringent region near the mid-plane ($y/H \approx 0.5$), which is anisotropic and mechanically weaker. As the crystallisation progresses, the birefringence fades away under small deformations in the linear regime [29], however, large deformations lead to crack formation, propagation, and eventually fracture in the birefringent regions [29]. These structural transitions are fairly consistent for matrix-dominated systems with filler concentrations up to $\Phi \approx 0.16$ (Fig. S4 and polarized microscopy videos in the SM [69]). The confined F127 matrix around SiO_2 fillers forms a thin anisotropic region where the material fractures ($\sim 25\text{--}80\text{ }\mu\text{m}$, Figs. 6(d) and S4 in the SM [69]).

The progression from affine flow to banding and fracture subtly differs from the original description of the slip-stick mechanism at the micellar level [79,80]. Instead, slip-slide-heal cycles take place at the fracture plane for matrix-dominated systems [29]. The fracture regime coincides with characteristic fingerprints in bulk properties. The matrix-dominated systems exhibit characteristic yielding features, including a G'' overshoot [71,72,81] and nonlinearities, $I_{3/1}$, at medium strain amplitudes that correspond to banding. While A_d values reach a plateau coinciding with fracture [Fig. 2(f)]. The filler concentration below this experimental threshold ($\Phi \sim 0.31$) does not change the yielding pathway, but it has an impact on material strength and energy dissipation. As Φ increases from 0 to ~ 0.31 , $G''_{\text{max}}/G''_{\text{LVR}}$ and γ_{nl} decrease, while ϕ_{LVR} and $A_{d,\text{max}}$ increase (Fig. 3). In short, although the rheological properties depend on filler concentration, the

deformation pathway remains matrix-dominated and qualitatively similar up to $\Phi \approx 0.31$.

2. Filler-dominated regime

In the filler-dominated regime ($\Phi > 0.31$), the linear regime shrinks and the characteristic G'' overshoot is lost [$G''_{\text{max}}/G''_{\text{LVR}} \rightarrow 1$, Figs. 2(c) and 3(a)]. The concentrated SiO_2 -F127 mixtures shift to a type-I strain softening behavior [71] which could be misinterpreted as homogeneous flow. However, microscopy shows that the material is highly heterogeneous and that localised motion of fillers takes place at very small strain amplitudes just before a crack forms within the linear region (Birefringence Video 3, SM). When the filler concentration reaches the percolation threshold, multiple light scattering at the contact points interferes with the birefringence analysis [Figs. 6(e)–6(h)]. The structural transitions or anisotropy of the confined F127 micelles in between the SiO_2 fillers cannot be quantified. Despite this limitation, fluorescence microscopy and PT results demonstrate that the deformation pathway is concentration dependent (Figs. 4 and 5).

The filler-dominated regime results in cracks and unyielded regions at strain amplitudes ($\gamma_0 \approx 0.1\%$) smaller than the onset of nonlinearity ($\gamma_{nl} \approx 0.21\%$ for $\Phi = 0.51$). Unlike for matrix-dominated systems, fracture location varies between test runs at high solids loading. Local events take place in random locations and propagate in multiple directions (see supporting videos 3–5 in the SM [69]). Although crack nucleation sites and subsequent fracture propagation vary slightly between runs, the critical strain and the overall failure behavior are consistent (see supporting videos in the SM [69]). These observations are also consistent with other heterogeneous materials where crack nucleation depends on the local

microstructure, but the overall failure behavior is maintained [82]. The displacement profile shows a step-like discontinuity for a representative run [at $\gamma_0 = 0.67\%$, Fig. 4(a)], where microscopy reveals a fracture [Fig. 4(e)]. The transient signals clearly split in two groups [Figs. 5(c) and 5(g)], corresponding to two distinct unyielded regions at $\gamma_0 = 1.4\%$ and $\gamma_0 = 9.8\%$ in the normalized displacement profiles [Figs. 5(d) and 5(h), respectively]. The tracers at the bottom move as a rigid body while the top region remains nearly static. Rigid body motion in highly filled suspensions in Newtonian fluids is associated with wall slip [83]. Here, slip occurs within the material when it fractures and not at the wall. A key aspect in this filler-dominated system is the absence of affine deformation and transition to a banding regime. Instead, the sample undergoes early fracture, bypassing the affine-banding-fracture transitions consistently observed in the matrix-dominated systems. Another characteristic feature is the significant energy dissipation from the start of the experiment at very small deformations [ϕ and A_d trends, Figs. 2(e), 2(f), 3(c) and 3(d)], which is consistent with prior work [30] and microscopy results [Figs. 6(e)–6(h)]. The area of the deltoids in the Cole-Cole plots reaches a peak [$A_{d,max}$, Fig. 3(d)] that coincides with the deltoid inversion (Fig. S2(c) in the SM [69]). The deltoid area plateaus around the maximum, and then it consistently decreases with increasing strain amplitude [A_d , Fig. 2(f)], owing to irreversible plastic and flow like contributions [30,42,84].

The intermediate regime ($\Phi \approx 0.3 - 0.4$) corresponds to a transition where particle-particle interaction becomes increasingly important. As a result, deformation becomes increasingly heterogeneous with stress localization controlled by local microstructure evolution, which ultimately leads to yielding or crack initiation, or a combination of both.

C. Reconciling bulk measures and micro-scale dynamics

Bringing together the key findings from bulk measurements and local observations, this study provides a map to establish the boundaries between three distinct deformation regimes: affine, banding, and fracture. The map presents the extent of deformation against filler concentration (γ_0 vs Φ , Fig. 7), which serves as an analog to phase diagrams widely used for steady shear and extensional flows. For example, shear-banding diagrams for wormlike micelles obtained using Rheo-NMR [85], flow state maps for simple yield stress fluids [35], and yielding maps for thixotropic suspensions [86]. Similarly, a recent study on adhesive granular suspensions under oscillatory shear proposed a yielding phase diagram linking energy dissipation to interparticle interactions [87].

Here, the map provides a generalized framework that systematically connects bulk rheological metrics to regime classification in filled yield stress matrices based on rheo-microscopy findings (Fig. 7). A sharp boundary exists between $\Phi = 0.31$ and 0.41 . At $\Phi \lesssim 0.31$, SiO₂-F127-water mixtures follow an affine-banding-fracture pathway, however, at $\Phi \gtrsim 0.41$ the composites bypass affine deformation and banding entirely. These distinctive deformation pathways at the microscale explain the bulk rheological metrics in the filler-dominated regime: the absence of the G'' overshoot, greater initial energy dissipation and decline in A_d , and higher

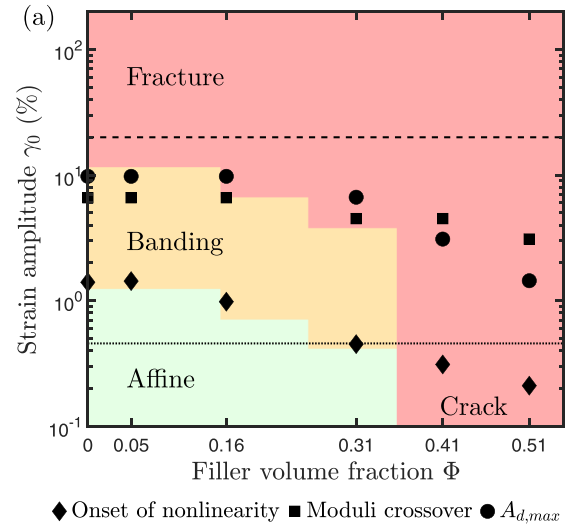


FIG. 7. Strain-concentration regime map for F127 gels with spherical silica fillers, summarizing deformation behavior across three regimes identified from bulk rheology and particle tracking (PT): green for affine deformation, yellow for shear banding, and red for fracture. Black diamond symbols ($\diamond$) mark the onset of nonlinearity (γ_{nl}) while black squares ($\blacksquare$) denote the flow strain ($G' = G''$). The horizontal dotted line marks the lower ($\gamma_0 = 0.46\%$) limit that can be quantified using particle tracking, corresponding to a displacement of $\approx 10 \mu\text{m}$. The dashed line marks the upper boundary ($\gamma_0 \approx 20\%$) that can be measured before the tracers leave the field of view at large amplitudes.

ϕ_{LVR} values that follow nonmonotonic trends due to irreversible events that lead to continued microstructural damage. The transition boundary in this map above $\Phi = 0.31$ is consistent with the observed trends for graphite-F127 formulations based on bulk measurements [30].

D. The importance of yielding pathways in direct ink writing applications and beyond

Bridging fundamentals to 3D printing applications, the results also show that the model DIW formulations (F127-SiO₂) match the observed bulk rheology and shape fidelity trends for analog systems [30]. Increasing the filler concentration (Φ , up to 0.51) improves the material strength by roughly an order of magnitude [G'_{LVR} , Fig. 8(a)], but it also leads to the loss of printing accuracy [Fig. 8(d)]. Additional criteria must be considered to assess printability [30,58]. A flowability map proposed in prior work [30] can capture the transition of yielding pathways with increasing solids loading [Fig. 8(b)]. This map shows a gradual shift from an abrupt to a gradual yielding transition with increased energy dissipation when filler concentration increases. The trends captured in the flowability map [Fig. 8(b)] are in agreement with the deformation regime phase diagram (Fig. 7). Both maps explain the loss of printing resolution due to increased and irreversible energy dissipation. Low- Φ matrix-dominated mixtures result in better shape fidelity [Fig. 8(c)] than the high- Φ filler-dominated composites [Fig. 8(d)].

Our rheo-microscopy results show that what we described as abrupt flowability means that the material is able to deform

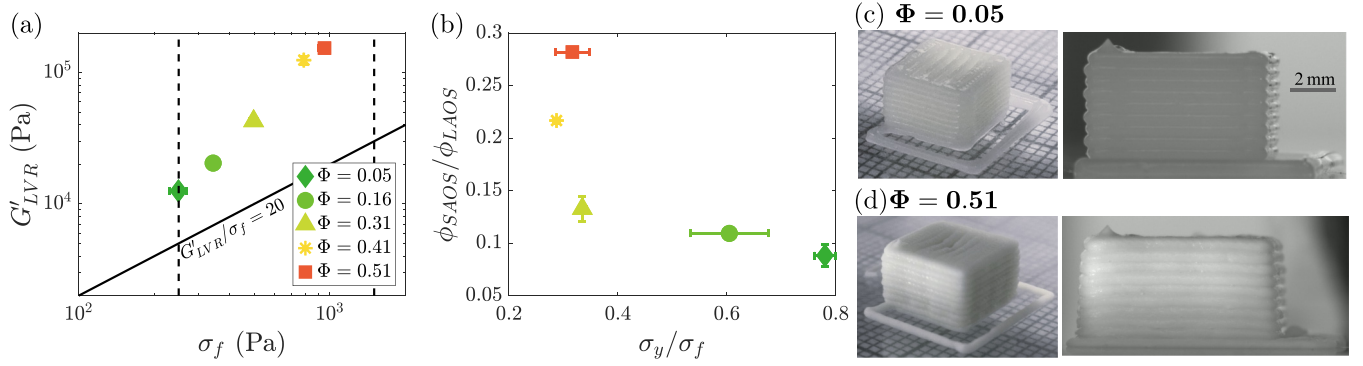


FIG. 8. (a), (b) Printability maps for F127 gel with SiO_2 fillers: (a) material strength map and (b) flowability map (dissipation ratio parameter $\frac{\phi_{\text{SAOS}}}{\phi_{\text{LAOS}}}$ vs stress ratio parameter $\frac{\sigma_y}{\sigma_f}$). The map in (a) indicates an increase in material strength with increasing volume fraction, quantified using the storage modulus in the linear viscoelastic region (G'_{LVR}) and the flow stress (σ_f), i.e., the stress amplitude at the moduli crossover ($G' = G''$). The map in (b) highlights a decrease in flowability, which is quantified using the dissipation ratio parameter ($\phi_{\text{SAOS}}/\phi_{\text{LAOS}}$) and the stress ratio parameter (σ_y/σ_f). Here, ϕ_{SAOS} and ϕ_{LAOS} are calculated at $\gamma_0 = 0.02\%$ and 100% , respectively. σ_y is the stress at the yield point, i.e., the onset of nonlinearity, $I_{3/1} > 0.01$. SiO_2 -F127 composites evolve from an abrupt ($\frac{\sigma_y}{\sigma_f} \rightarrow 1$, and $\frac{\phi_{\text{SAOS}}}{\phi_{\text{LAOS}}} \rightarrow 0$) to a gradual ($\frac{\sigma_y}{\sigma_f} \rightarrow 0$, and $\frac{\phi_{\text{SAOS}}}{\phi_{\text{LAOS}}} \uparrow$) yielding transition with increased energy dissipation at increasing volume fractions (Φ). (c), (d) Images showing representative 3D-printed parts using formulations with different Φ values. Panel (c) qualitatively shows good print quality obtained using the matrix-dominated formulation ($\Phi = 0.05$). The same printed parts produced with the filler-dominated system [$\Phi = 0.51$, (d)] exhibit obvious defects, such as a collapse in the middle of the 3D lattice and a loss of filament resolution.

elastically from affine to banding without microscopic evidence of plastic deformation until it fractures. While gradual flowability corresponds to localized plastic events that lead to catastrophic failure within our experimental timescales. Resembling the strength-toughness trade-off in composites microstructure engineering [88], here the fillers increase the strength of the composite but they also undermine the toughness of the matrix at high solids loading, which compromises printing outcomes.

IV. CONCLUSIONS

Bringing together bulk measurements and quantitate direct visualization, we have addressed the central question on this study by demonstrating that yielding and fracture coexist within concentrated suspensions in a hard gel matrix, and that the deformation pathway directly depends on the solids loading. Above certain concentration threshold ($\Phi \sim 31$ vol%), the matrix is confined within the fillers, leading to local plastic events, early onset of nonlinearities, and increased energy dissipation even at the smallest strain amplitudes. The linear regime shrinks with increasing solids loading. Cracks propagate and fracture the material into solid like and yielded regions as the applied strain increases. The fillers improve the strength but undermine the toughness of the composite network, which loses its ability to deform elastically. As solids loading increases, the material fails to recover its original structure after the high-shear extrusion process, leading to the observed loss of print fidelity.

This approach elucidates underlying deformation mechanisms and enables the reconciliation of bulk metrics with local observations. These findings can be generalized, for example, a growth and peak in the area of the deltoids in the sequence of physical processes framework at intermediate strain ampli-

tudes signifies sustained banding, while an early maximum followed by decline indicates fracture.

The significance of these findings lies in our ability to bridge the gap between fundamentals and applications by providing a deformation regime map that captures distinctive yielding pathways for concentrated suspensions in yield stress matrices. This map serves as a tool to identify strain-concentration combinations that minimize failure during flow of soft materials. The findings also open avenues in microstructure engineering of soft matter to achieve a compromise between strength and toughness. This is critical for applications such as bioprinting and direct ink writing of advanced functional materials.

ACKNOWLEDGMENTS

This work was supported by a UKRI Future Leaders Fellowship No. MR/V021117/1. The authors acknowledge Prof. S. Rogers for providing the SPP Freeware package to carry out the SPP analysis.

E.G.T.; data curation: R.A., E.G.T.; formal analysis: R.A., E.G.T.; funding acquisition: E.G.T.; investigation: E.G.T., P.S., R.A.; methodology: E.G.T., R.A.; project administration: E.G.T.; resources: E.G.T.; software: R.A., E.G.T.; supervision: E.G.T.; writing original draft: R.A., E.G.T.; editing: E.G.T., P.S.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

- [1] D. A. Rau, C. B. Williams, and M. J. Bortner, Rheology and printability: A survey of critical relationships for direct ink write materials design, *Prog. Mater. Sci.* **140**, 101188 (2023).
- [2] P. Fischer and E. J. Windhab, Rheology of food materials, *Curr. Opin. Colloid Interface Sci.* **16**, 36 (2011).
- [3] R. H. Ewoldt, P. Winter, J. Maxey, and G. H. McKinley, Large amplitude oscillatory shear of pseudoplastic and elastoviscoplastic materials, *Rheol. Acta* **49**, 191 (2010).
- [4] R. Kostynick, H. Matinpour, S. Pradeep, S. Haber, A. Sauret, E. Meiburg, T. Dunne, P. Arratia, and D. Jerolmack, Rheology of debris flow materials is controlled by the distance from jamming, *Proc. Natl. Acad. Sci. USA* **119**, e2209109119 (2022).
- [5] D. Bonn, M. M. Denn, L. Berthier, T. Divoux, and S. Manneville, Yield stress materials in soft condensed matter, *Rev. Mod. Phys.* **89**, 035005 (2017).
- [6] H. A. Barnes, The yield stress—a review or ‘*παντα ρει*’—everything flows? *J. Non-Newtonian Fluid Mech.* **81**, 133 (1999).
- [7] M. Kamkar, R. Salehiyan, T. B. Goudoulas, M. Abbasi, C. Saengow, E. Erfanian, S. Sadeghi, G. Natale, S. A. Rogers, A. J. Giacomini, *et al.*, Large amplitude oscillatory shear flow: Microstructural assessment of polymeric systems, *Prog. Polym. Sci.* **132**, 101580 (2022).
- [8] K. Hyun, M. Wilhelm, C. O. Klein, K. S. Cho, J. G. Nam, K. H. Ahn, S. J. Lee, R. H. Ewoldt, and G. H. McKinley, A review of nonlinear oscillatory shear tests: Analysis and application of large amplitude oscillatory shear (LAOS), *Prog. Polym. Sci.* **36**, 1697 (2011).
- [9] E. C. Bingham, *Fluidity and Plasticity* (McGraw-Hill Book Company, Incorporated, New York, 1922).
- [10] P. Saramito, A new elastoviscoplastic model based on the Herschel–Bulkley viscoplastic model, *J. Non-Newtonian Fluid Mech.* **158**, 154 (2009).
- [11] K. Kamani, G. J. Donley, and S. A. Rogers, Unification of the rheological physics of yield stress fluids, *Phys. Rev. Lett.* **126**, 218002 (2021).
- [12] K. M. Kamani and S. A. Rogers, Brittle and ductile yielding in soft materials, *Proc. Natl. Acad. Sci. USA* **121**, e2401409121 (2024).
- [13] S. Aime, D. Truzzolillo, D. J. Pine, L. Ramos, and L. Cipelletti, A unified state diagram for the yielding transition of soft colloids, *Nat. Phys.* **19**, 1673 (2023).
- [14] A. Corker, H. C.-H. Ng, R. J. Poole, and E. García-Tuñón, 3D printing with 2D colloids: Designing rheology protocols to predict “printability” of soft-materials, *Soft Matter* **15**, 1444 (2019).
- [15] M. Maillard, S. Fournier, E. García-Tuñón, and E.-J. Courtial, Advances and obstacles to unify the concept of “printability” in ceramics direct ink writing, *Open Ceram.* **23**, 100808 (2025).
- [16] B. Ling, R. Agrawal, D. He, R. J. Poole, A. I. Cooper, M. Liu, and E. García-Tuñón, 3D-printed porous organic cages for gas filtration: Fabrication and flow simulations, *Adv. Funct. Mater.* **34**, 2405320 (2024).
- [17] B. Ling, H. Wang, P. Yang, R. Agrawal, A. R. Neale, L. J. Hardwick, T. Hasell, and E. García-Tuñón, 3D-printed sulfur-derived polymers with controlled architectures for lithium-sulfur batteries, *Adv. Funct. Mater.* e12927 (2026).
- [18] E. García-Tuñón, R. Agrawal, B. Ling, and D. J. Dennis, Fourier-transform rheology and printability maps of complex fluids for three-dimensional printing, *Phys. Fluids* **35**, 017113 (2023).
- [19] E. García-Tuñón, E. Feilden, H. Zheng, E. D’Elia, A. Leong, and E. Saiz, Graphene oxide: An all-in-one processing additive for 3D printing, *ACS Appl. Mater. Interfaces* **9**, 32977 (2017).
- [20] R. Suntornnond, E. Y. S. Tan, J. An, and C. K. Chua, A highly printable and biocompatible hydrogel composite for direct printing of soft and perfusable vasculature-like structures, *Sci. Rep.* **7**, 16902 (2017).
- [21] Y. He, F. Yang, H. Zhao, Q. Gao, B. Xia, and J. Fu, Research on the printability of hydrogels in 3D bioprinting, *Sci. Rep.* **6**, 29977 (2016).
- [22] Z. L. Li, S. Zhou, E. Saiz, and R. Malik, Ink formulation in direct ink writing of ceramics: A meta-analysis, *J. Eur. Ceram. Soc.* **44**, 6777 (2024).
- [23] K. Mortensen, Phase behaviour of poly(ethylene oxide)-poly(propylene oxide)-poly(ethylene oxide) triblock-copolymer dissolved in water, *Europhys. Lett.* **19**, 599 (1992).
- [24] G. E. Newby, I. W. Hamley, S. M. King, C. M. Martin, and N. J. Terrill, Structure, rheology and shear alignment of Pluronic block copolymer mixtures, *J. Colloid Interface Sci.* **329**, 54 (2009).
- [25] K. Suman, S. Sourav, and Y. M. Joshi, Rheological signatures of gel-glass transition and a revised phase diagram of an aqueous triblock copolymer solution of Pluronic F127, *Phys. Fluids* **33**, 073610 (2021).
- [26] N. A. Di Spirito, N. Grizzuti, and R. Pasquino, Self-assembly of Pluronics: A critical review and relevant applications, *Phys. Fluids* **36**, 111302 (2024).
- [27] A. Oshiro, D. C. da Silva, J. C. de Mello, V. W. de Moraes, L. P. Cavalcanti, M. K. Franco, M. I. Alkschbirs, L. F. Fraceto, F. Yokaichiya, T. Rodrigues, *et al.*, Pluronics F-127/L-81 binary hydrogels as drug-delivery systems: influence of physicochemical aspects on release kinetics and cytotoxicity, *Langmuir* **30**, 13689 (2014).
- [28] M. Jalaal, G. Cottrell, N. Balmforth, and B. Stoeber, On the rheology of Pluronic F127 aqueous solutions, *J. Rheol.* **61**, 139 (2017).
- [29] R. Agrawal, P. T. Spicer, and E. García-Tuñón, Connecting bulk rheology, structural transitions and heterogeneous flow in Pluronic F127 micellar cubic liquid crystals using rheo-microscopy, *J. Colloid Interface Sci.* **699**, 138226 (2025).
- [30] R. Agrawal and E. García-Tuñón, Interplay between yielding, ‘recovery’, and strength of yield stress fluids for direct ink writing: New insights from oscillatory rheology, *Soft Matter* **20**, 7429 (2024).
- [31] K. Hyun, J. G. Nam, M. Wilhelm, K. H. Ahn, and S. J. Lee, Large amplitude oscillatory shear behavior of PEO-PPO-PEO triblock copolymer solutions, *Rheol. Acta* **45**, 239 (2006).
- [32] E. Feilden, E. G.-T. Blanca, F. Giuliani, E. Saiz, and L. Vandeperre, Robocasting of structural ceramic parts with hydrogel inks, *J. Eur. Ceram. Soc.* **36**, 2525 (2016).
- [33] E. Feilden, C. Ferraro, Q. Zhang, E. García-Tuñón, E. D’Elia, F. Giuliani, L. Vandeperre, and E. Saiz, 3D printing bioinspired ceramic composites, *Sci. Rep.* **7**, 13759 (2017).
- [34] P. Schall, D. A. Weitz, and F. Spaepen, Structural rearrangements that govern flow in colloidal glasses, *Science* **318**, 1895 (2007).

- [35] T. Divoux, D. Tamarii, C. Barentin, and S. Manneville, Transient shear banding in a simple yield stress fluid, *Phys. Rev. Lett.* **104**, 208301 (2010).
- [36] R. W. Mair and P. T. Callaghan, Observation of shear banding in worm-like micelles by NMR velocity imaging, *Europhys. Lett.* **36**, 719 (1996).
- [37] G. J. Donley, S. Narayanan, M. A. Wade, J. D. Park, R. L. Leheny, J. L. Harden, and S. A. Rogers, Investigation of the yielding transition in concentrated colloidal systems via rheo-XPCS, *Proc. Natl. Acad. Sci. USA* **120**, e2215517120 (2023).
- [38] H. J. Barlow, J. O. Cochran, and S. M. Fielding, Ductile and brittle yielding in thermal and athermal amorphous materials, *Phys. Rev. Lett.* **125**, 168003 (2020).
- [39] M. Ozawa, L. Berthier, G. Biroli, A. Rosso, and G. Tarjus, Random critical point separates brittle and ductile yielding transitions in amorphous materials, *Proc. Natl. Acad. Sci. USA* **115**, 6656 (2018).
- [40] T. Divoux, M. A. Fardin, S. Manneville, and S. Lerouge, Shear banding of complex fluids, *Annu. Rev. Fluid Mech.* **48**, 81 (2016).
- [41] J. Goyon, A. Colin, G. Ovarlez, A. Ajdari, and L. Bocquet, Spatial cooperativity in soft glassy flows, *Nature (London)* **454**, 84 (2008).
- [42] S. A. Rogers, In search of physical meaning: Defining transient parameters for nonlinear viscoelasticity, *Rheol. Acta* **56**, 501 (2017).
- [43] M. Fuchs and M. E. Cates, Theory of nonlinear rheology and yielding of dense colloidal suspensions, *Phys. Rev. Lett.* **89**, 248304 (2002).
- [44] P. Sollich, F. Lequeux, P. Hébraud, and M. E. Cates, Rheology of soft glassy materials, *Phys. Rev. Lett.* **78**, 2020 (1997).
- [45] J. M. Brader, T. Voigtmann, M. Fuchs, R. G. Larson, and M. E. Cates, Glass rheology: From mode-coupling theory to a dynamical yield criterion, *Proc. Natl. Acad. Sci. USA* **106**, 15186 (2009).
- [46] S. Fielding, M. Cates, and P. Sollich, Shear banding, aging and noise dynamics in soft glassy materials, *Soft Matter* **5**, 2378 (2009).
- [47] P. Sollich, Challenges in the rheology of glasses, *arXiv:2401.16409*.
- [48] J. O. Cochran, G. L. Callaghan, M. J. G. Caven, and S. M. Fielding, Slow fatigue and highly delayed yielding via shear banding in oscillatory shear, *Phys. Rev. Lett.* **132**, 168202 (2024).
- [49] A. Nicolas, E. E. Ferrero, K. Martens, and J.-L. Barrat, Deformation and flow of amorphous solids: Insights from elastoplastic models, *Rev. Mod. Phys.* **90**, 045006 (2018).
- [50] J. Lin, E. Lerner, A. Rosso, and M. Wyart, Scaling description of the yielding transition in soft amorphous solids at zero temperature, *Proc. Natl. Acad. Sci. USA* **111**, 14382 (2014).
- [51] K. Martens, L. Bocquet, and J.-L. Barrat, Spontaneous formation of permanent shear bands in a mesoscopic model of flowing disordered matter, *Soft Matter* **8**, 4197 (2012).
- [52] T. Jocteur, S. Figueiredo, K. Martens, E. Bertin, and R. Mari, Yielding is an absorbing phase transition with vanishing critical fluctuations, *Phys. Rev. Lett.* **132**, 268203 (2024).
- [53] B. P. Bhowmik and C. Ness, Scaling description of frictionless dense suspensions under inhomogeneous flow, *Phys. Rev. Lett.* **132**, 118203 (2024).
- [54] C. Ness and S. M. Fielding, Nonmonotonic constitutive curves and shear banding in dry and wet granular flows, *Phys. Rev. Lett.* **134**, 038201 (2025).
- [55] J. A. Lewis, Direct ink writing of 3D functional materials, *Adv. Funct. Mater.* **16**, 2193 (2006).
- [56] M. Saadi, A. Maguire, N. T. Pottackal, M. S. H. Thakur, M. M. Ikram, A. J. Hart, P. M. Ajayan, and M. M. Rahman, Direct ink writing: A 3D printing technology for diverse materials, *Adv. Mater.* **34**, 2108855 (2022).
- [57] P. Wei, C. Cipriani, C.-M. Hsieh, K. Kamani, S. Rogers, and E. Pentzer, Go with the flow: Rheological requirements for direct ink write printability, *J. Appl. Phys.* **134**, 100701 (2023).
- [58] Y. Liu, M. Hildner, O. Roy, W. A. Van den Bogert, J. Lorenz, M. Desroches, K. Koppi, A. Shih, and R. G. Larson, On the selection of rheological tests for the prediction of 3D printability, *J. Rheol.* **67**, 791 (2023).
- [59] C. E. McNamee, K. Graf, H.-J. Butt, K. Higashitani, and M. Kappl, Interaction between a silica particle and the underside of a polymer monolayer at the air/water interface in the presence of an anionic surfactant, *Colloids Surf. A* **383**, 32 (2011).
- [60] L. C. Geonzon, M. Kobayashi, T. Sugimoto, and Y. Adachi, Study on the kinetics of adsorption of poly(ethylene oxide) onto a silica particle using optical tweezers and microfluidics, *Colloids Surf. A* **642**, 128691 (2022).
- [61] V. Trappe and D. A. Weitz, Scaling of the viscoelasticity of weakly attractive particles, *Phys. Rev. Lett.* **85**, 449 (2000).
- [62] E. Brown and H. M. Jaeger, Shear thickening in concentrated suspensions: Phenomenology, mechanisms and relations to jamming, *Rep. Prog. Phys.* **77**, 046602 (2014).
- [63] P. B. Laxton and J. C. Berg, Gel trapping of dense colloids, *J. Colloid Interface Sci.* **285**, 152 (2005).
- [64] R. H. Ewoldt, A. Hosoi, and G. H. McKinley, New measures for characterizing nonlinear viscoelasticity in large amplitude oscillatory shear, *J. Rheol.* **52**, 1427 (2008).
- [65] S. A. Rogers, B. M. Erwin, D. Vlassopoulos, and M. Cloitre, A sequence of physical processes determined and quantified in LAOS: Application to a yield stress fluid, *J. Rheol.* **55**, 435 (2011).
- [66] S. A. Rogers and M. P. Lettinga, A sequence of physical processes determined and quantified in large-amplitude oscillatory shear (LAOS): Application to theoretical nonlinear models, *J. Rheol.* **56**, 1 (2012).
- [67] J.-Y. Tinevez, N. Perry, J. Schindelin, G. M. Hoopes, G. D. Reynolds, E. Laplantine, S. Y. Bednarek, S. L. Shorte, and K. W. Eliceiri, TrackMate: An open and extensible platform for single-particle tracking, *Methods* **115**, 80 (2017).
- [68] D. Ershov, M.-S. Phan, J. W. Pyllänen, S. U. Rigaud, L. Le Blanc, A. Charles-Orszag, J. R. Conway, R. F. Laine, N. H. Roy, D. Bonazzi, *et al.*, TrackMate 7: integrating state-of-the-art segmentation algorithms into tracking pipelines, *Nat. Methods* **19**, 829 (2022).
- [69] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/d86k-mgq6> for additional visual items showing the diw system and 3D design, supporting Lissajous-Bowditch and Cole-Cole plots, SPP trajectories, birefringence mapping results, and supporting videos.
- [70] J. A. Lewis, J. E. Smay, J. Stuecker, and J. Cesarano, Direct ink writing of three-dimensional ceramic structures, *J. Am. Ceram.* **89**, 3599 (2006).

- [71] K. Hyun, S. H. Kim, K. H. Ahn, and S. J. Lee, Large amplitude oscillatory shear as a way to classify the complex fluids, *J. Non-Newtonian Fluid Mech.* **107**, 51 (2002).
- [72] G. J. Donley, P. K. Singh, A. Shetty, and S. A. Rogers, Elucidating the G'' overshoot in soft materials with a yield transition via a time-resolved experimental strain decomposition, *Proc. Natl. Acad. Sci. USA* **117**, 21945 (2020).
- [73] K. M. Kamani, G. J. Donley, R. Rao, A. M. Grillet, C. Roberts, A. Shetty, and S. A. Rogers, Understanding the transient large amplitude oscillatory shear behavior of yield stress fluids, *J. Rheol.* **67**, 331 (2023).
- [74] R. Agrawal, E. García-Tuñón, R. J. Poole, and C. P. Fonte, Features and limitations of recent elastoviscoplastic constitutive models under Large Amplitude Oscillatory Shear (LAOS), *J. Non-Newtonian Fluid Mech.* **338**, 105407 (2025).
- [75] A. Fall, N. Huang, F. Bertrand, G. Ovarlez, and D. Bonn, Shear thickening of cornstarch suspensions as a reentrant jamming transition, *Phys. Rev. Lett.* **100**, 018301 (2008).
- [76] J. F. Morris, Shear thickening of concentrated suspensions: Recent developments and relation to other phenomena, *Annu. Rev. Fluid Mech.* **52**, 121 (2020).
- [77] A. Rodríguez-Palomo, V. Lutz-Bueno, M. Guizar-Sicairos, R. Kádár, M. Andersson, and M. Liebi, Nanostructure and anisotropy of 3D printed lyotropic liquid crystals studied by scattering and birefringence imaging, *Addit. Manuf.* **47**, 102289 (2021).
- [78] N. Balsara, D. Perahia, C. Safinya, M. Tirrell, and T. Lodge, Birefringence detection of the order-to-disorder transition in block copolymer liquids, *Macromolecules* **25**, 3896 (1992).
- [79] I. W. Hamley, J. A. Pople, C. Booth, L. Derici, M. Impéror-Clerc, and P. Davidson, Shear-induced orientation of the body-centered-cubic phase in a diblock copolymer gel, *Phys. Rev. E* **58**, 7620 (1998).
- [80] C. R. López-Barrón, L. Porcar, A. P. Eberle, and N. J. Wagner, Dynamics of melting and recrystallization in a polymeric micellar crystal subjected to large amplitude oscillatory shear flow, *Phys. Rev. Lett.* **108**, 258301 (2012).
- [81] Y. H. Shim and S. A. Rogers, Understanding the yielding behavior of graphene oxide colloids via experimental strain decomposition, *Phys. Fluids* **35**, 063117 (2023).
- [82] D. Bonamy, S. Santucci, and L. Ponson, Crackling dynamics in material failure as the signature of a self-organized dynamic phase transition, *Phys. Rev. Lett.* **101**, 045501 (2008).
- [83] D. M. Kalyon and S. Aktaş, Factors affecting the rheology and processability of highly filled suspensions, *Annu. Rev. Chem. Biomol. Eng.* **5**, 229 (2014).
- [84] E. L. Jones, Z. Luo, R. Agrawal, S. Flynn, M. Carr, W. Sharratt, and E. García-Tuñón, Designing yield stress fluids for advanced materials processing using derivatives of pH-responsive branched co-polymer surfactants, *Soft Matter* **21**, 4822 (2025).
- [85] M. Lopez-Gonzalez, W. Holmes, and P. Callaghan, Rheo-NMR phenomena of wormlike micelles, *Soft Matter* **2**, 855 (2006).
- [86] P. C. F. Møller, S. Rodts, M. A. J. Michels, and D. Bonn, Shear banding and yield stress in soft glassy materials, *Phys. Rev. E* **77**, 041507 (2008).
- [87] S. Chattopadhyay, S. Nagaraja, and S. Majumdar, Effect of adhesive interaction on strain stiffening and dissipation in granular gels undergoing yielding, *Commun. Phys.* **5**, 126 (2022).
- [88] O. Gavalda-Diaz, E. Saiz, J. Chevalier, and F. Bouville, Toughening of ceramics and ceramic composites through microstructure engineering: A review, *Int. Mater. Rev.* **70**, 3 (2025).