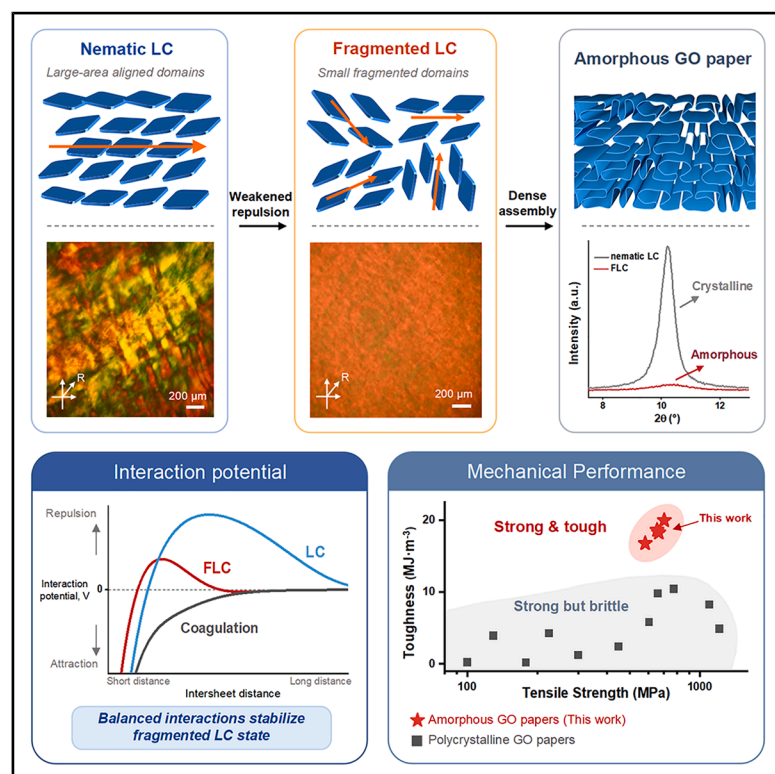


Fragmented liquid crystals of graphene oxide

Graphical abstract



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In brief

Shen et al. identify fragmented liquid crystals in graphene oxide as an intermediate state between ordinary nematic liquid crystals and aggregation. Enabled by balanced inter-sheet interactions, large liquid-crystal domains fragment into small, sheet-scale regions while retaining fluidity and processability. This frustrated order provides an unexpected route to compact amorphous architectures distinct from conventional crystalline-oriented liquid-crystal assembly, suppressing large structural defects and combining high strength with toughness.

Highlights

- Fragmented liquid crystals bridge nematic order and aggregation in GO
- Balanced interactions preserve fluidity while fragmenting LC domains
- GO FLCs assemble into compact amorphous architectures
- Amorphous GO papers combine enhanced strength and toughness



Discovery

A new material or phenomena

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Article

Fragmented liquid crystals of graphene oxide

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PROGRESS AND POTENTIAL Liquid crystals (LCs) formed by repulsive colloids are widely used to organize anisotropic building blocks into ordered materials. When repulsion is weakened, such systems are generally expected to collapse into coagulates or gels, losing structural order or processability. Here, we identify an intermediate LC mesophase of graphene oxide (GO), termed fragmented liquid crystal (FLC), between nematic LCs and coagulates/gels. FLC features fragmented LC domains down to single-entity scale while retaining fluidity. It arises from the balance between weakened electrostatic repulsion and transient inter-sheet attraction, maintaining the single-sheet dispersed state while forming transient networks. This finding reveals a previously unrecognized intermediate mesophase in GO colloidal LCs and offers a route to regulate orientational order down to the single-entity scale. More broadly, these findings suggest that analogous FLC states may be accessible in other highly anisotropic nanosheet colloids when interparticle interactions and dispersion stability can be appropriately controlled.

Beyond being a physical state, FLC provides a useful processing window. By disrupting large-scale alignment while preserving dense packing, GO FLCs assemble into compact amorphous papers, unlike the brittle polycrystalline laminates from ordinary nematic LCs. This amorphous yet densely packed assembly suppresses major structural defects, enabling simultaneous improvements in strength and toughness. Distinct from the prevailing crystalline-oriented trend, FLC provides an alternative route to designing amorphous structures of flexible 2D sheets. Since this state can be accessed by simple ionic-strength modulation and is compatible with solution processing, it may guide the design of mechanically reliable materials from GO and other anisotropic nanosheet colloids.

SUMMARY

Repulsive colloids can form liquid crystals (LCs) that combine fluidity and structural order. Screening repulsion leads to abrupt LC collapse, causing either loss of fluidity by gelation or frustrated structural order by coagulation. However, the evolution of liquid crystallinity before the transition from overall repulsion to attraction remains unclear. Here, we find an intermediate LC state of graphene oxide (GO), named fragmented LC (FLC). FLC features fragmented domains down to single-entity size but keeps fluidity, unlike gels and coagulates. In FLC, balanced interactions maintain the single-layer dispersed state of GO and trigger transient networks. GO FLCs surprisingly serve as a peculiar processing state toward compact amorphous structures with high strength and toughness, beyond brittle polycrystalline structures assembled from ordinary nematic LCs. Our findings enable precisely modulating LC directors down to the molecular-size limit and provide a path to designing amorphous structures of flexible 2D sheets beyond the prevailing crystalline-oriented trend.

INTRODUCTION

Liquid crystal (LC) is the fourth essential matter state and features both fluidity of liquid and structural orders as crystals.^{1,2} Repulsive colloids and giant molecules with high anisotropy can form lyotropic LCs,^{3,4} extending from one-dimensional cellulose nanocrystals⁵ to two-dimensional (2D) colloidal clays⁶ and 2D molecular graphene oxide (GO).⁷ Beyond serving as a model 2D colloidal mesogen, GO is also a scalable and solution-processable precursor to graphene-based macroscopic materials, including films, fibers, and aerogels, with applications in separation, energy-storage, thermal management, and structural materials.^{8–10} In the framework of Onsager theory, dominant repulsion drives the alignment of anisotropic entities. The resulting gain in translational entropy compensates the loss of orientational entropy, enabling the formation of LCs as an entropy-favored spontaneous process.¹¹ Complex interactions between mesogenic units enable the formation of rich LCs, such as nematic,¹² columnar,¹³ lamellar,¹⁴ chiral,¹⁵ and cubic¹⁶ phases, continuously enriching the understanding of fluid physics and matter organization. LC order in fluids provides a practical route to assemble solids with targeted structures.⁸ For example, colloidal LCs have been processed to high-performance assemblies with superior functions, such as fibers of graphene, carbon nanotubes, and cellulose by wet-spinning^{9,17,18} and their strong films by fluid casting.¹⁹ Colloidal LCs are highly tunable and responsive.²⁰ Precisely modulating LC orders has become an efficient methodology to tune the fluid-to-solid assembly process and regulate structures and properties of materials.^{21–23}

Repulsion dominates the formation of colloidal LCs. Weakening repulsion can lead to the collapse of LC orders.^{24,25} Previous studies have identified two structural collapse endpoints of LC orders: coagulations at low concentration and gels at high concentration. LCs are defined by the coexistence of orientational order and fluidity. As repulsion diminishes, mesogenic units come into direct contact, thereby breaking this balance. The system typically evolves either toward irreversible coagulation with uncontrolled aggregation^{26,27} or toward gelation where a space-spanning network suppresses fluidity.^{28,29} In both cases, fluidity is lost and long-range orientational correlations deteriorate. The ordered gelation of LC has been widely used to wet-spun fibers with ordered structure and high performances.^{18,30–32} Beyond these two terminations, theoretic analysis predicts that LC should survive as the weakening repulsion still overwhelms the attraction before the critical transition point.¹² However, the evolution of frustrating LC orders before their final collapse has long been ignored and the potential of this intermediate transition state in material structural design still waits to be explored.

Here, we identify an intermediate frustrated mesophase, termed as fragmented LC (FLC), in 2D colloidal LCs with weakened repulsion. This FLC state is distinct from ordinary nematic LCs with strong repulsion and coagulations or gels with strong attraction. We exploit the intermediate FLC state to achieve compact amorphous assembled graphene materials. Moderate monovalent ions or heating weaken the repulsion and turn the large-area nematic GO LC to FLC with small LC domain down to the lateral size of GO sheets. Balanced interactions

keep GO single-layer dispersed in the FLC state. At the same time, transient inter-sheet connectivity forms a dynamic network of fragmented domains while preserving the fluidity. GO FLC surprisingly serves as a peculiar processing state to assemble amorphous but compact graphene papers with both outstanding strength and toughness, distinct from stiff and brittle polycrystalline structures assembled from large-area nematic LCs. FLC enables precise modulation of LC directors and offers a distinctive processing window for assembling materials. The amorphous GO FLC papers provide an alternative route to designing amorphous structures of flexible 2D sheets, diverging from the prevailing crystalline-oriented trend.

RESULTS

GO FLC and its characteristics

We tracked the evolution of LCs with increasing ionic strength. To avoid crosslinking effects, the ionic strength was adjusted using monovalent ions (e.g., potassium chloride, KCl) rather than multivalent salts. Increasing ionic strength can reduce the electrostatic repulsion by compressing the electric double layer (EDL) and the screening surface potential.³³ In the absence of KCl, nematic GO LC shows colorful large-area nematic LC regions between crossed polarizers (Figure 1a₁) and exhibits homogeneity at both macroscopic and microscopic scales (Figure S1), usually as a good processing state for macroscopic assembled materials such as fibers and films.⁸ At high KCl concentration (C_{KCl}) of 100 mM (mmol/L), attraction overcomes repulsion and leads to the gelation of GO sheets (Figure 1a₄). The extreme gelation state of GO loses fluidity and breaks into smaller discontinuous coagulation granules, completely losing fluid processability (Figure 1b₃).

At intermediate C_{KCl} (typically 5–20 mM), a distinct LC mesophase emerges. The first notable observation is that large LC domains gradually break up into smaller domains and the fragmented LC domains are even too small to be distinguished by naked eyes at 15 mM of C_{KCl} (Figures 1a₂ and 1a₃). For this characteristic, we name this intermediate state as FLCs. Similar to GO LCs without KCl, GO FLCs still have macroscopic uniform appearance (Figure S1B) and show homogeneity without aggregation at microscopic scale (Figure S1E). GO FLC is a long-term stable intermediate state, showing no detectable change in its homogeneity and FLC structure during a 1-week observation period (Figure S2). In contrast with the extreme coagulations, GO FLCs retain fluidity and can flow homogeneously and continuously under external forces (Figures 1b₁ and 1b₂). The LC textures of GO FLCs still maintain during the extrusion and subsequent flow (Figure S3). GO FLCs have higher viscosity (950 Pa·s) than ordinary nematic GO LCs (177 Pa·s) (Figure S14A) and can thus maintain certain shape after extrusion. Beyond potassium ions, GO FLC is a general state that can be triggered by other monovalent salts or by moderate heating, both of which reduce the electrostatic repulsion between GO sheets (Figure S4). The FLC state was also reproduced in an ~800 mL batch at a final KCl concentration of ~15 mM, confirming that FLC formation is not restricted to small-volume preparations (Figure S5).

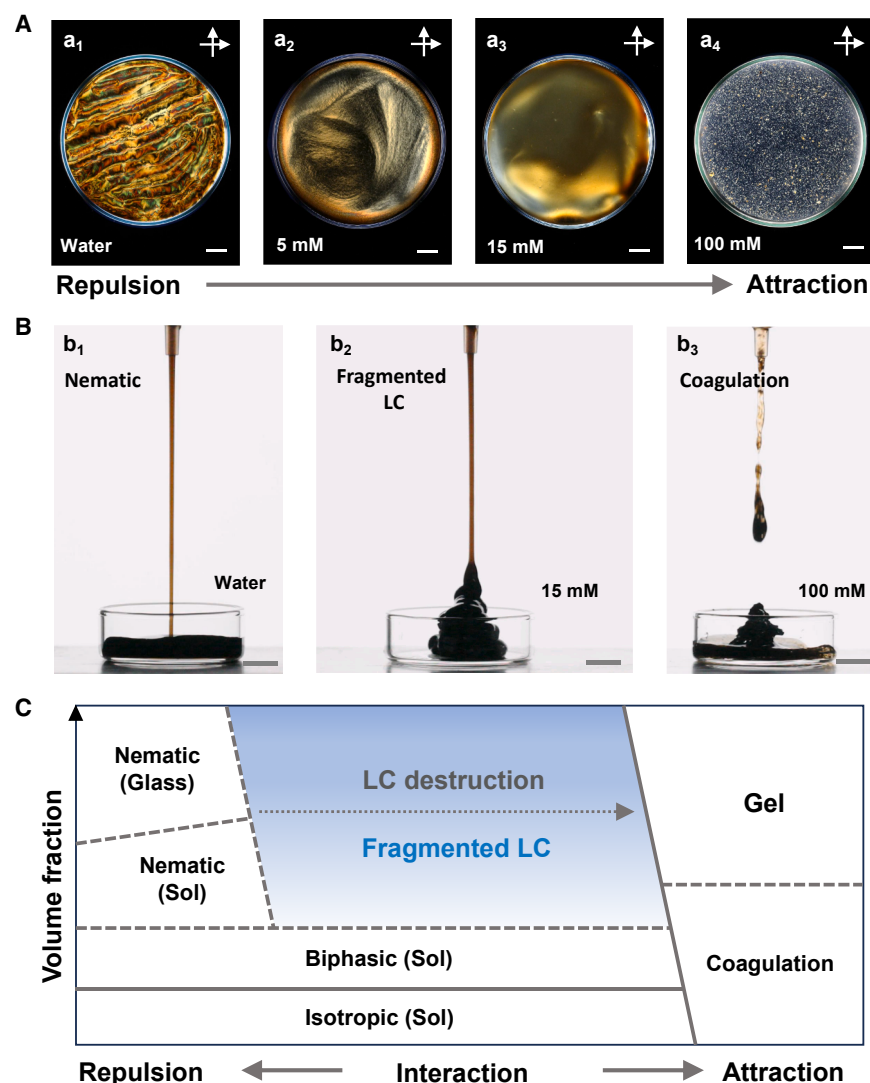


Figure 1. GO FLC and its characteristics

(A) Optical image under crossed polarizers showing the evolution of GO dispersions with increasing C_{KCl} , where large-area nematic LC domains gradually fragment into smaller LC regions and eventually form coagulations. Increasing ionic strength can reduce the electrostatic repulsion by compressing the EDL and the screening surface potential.

At 15 mM C_{KCl} , the FLC domains are too small to be distinguished in the large area image.

(B) Flow behaviors of GO dispersions at different C_{KCl} . The GO dispersion at 15 mM, containing FLC domains, exhibits uniform and continuous flow, in contrast to the coagulated samples.

(C) Phase diagram of GO dispersions including different LC phases (isotropic, biphasic, and nematic) and arrested states (sol, glass, and gel/coagulation). Further explanations of these phases are provided in the [Note S1](#).

The GO LC state featuring fragmented small LC domains, observed at intermediate KCl concentrations, is regarded as an intermediate LC processing state with balanced repulsion and attraction between nematic LCs and coagulations/gels, referred to as FLCs. Scale bars, 1 cm (A and B).

Based on this observation, we revisit the organization state of GO dispersions in relation with intermolecular interactions and solid content ([Figure 1C](#)). In aqueous systems with very low ionic concentration (e.g., pure water, $\sim 10^{-3}$ mM), GO sheets keep strongly repulsive and form stable nematic LCs. At the opposite extreme of high ionic concentrations, the shielded electrostatic repulsion is outweighed by the van der Waals attraction and hydrogen bonds, leading to coagulation or gelation.³³ Our observation reveals that there is no sharp boundary between these two extremes. The FLC state, characterized by small FLC regions, represents an intermediate regime where repulsion and attraction are balanced ([Figure 1C](#)). This intermediate state retains both fluidity and structural order, constituting a previously unexplored LC processing state.

Track the formation of GO FLC

The fragmentation of LC domains in FLC is confirmed by polarized optical microscope (POM) and scanning electron

microscope (SEM) ([Figures 2A](#) and [2B](#)). In POM images, FLCs exhibit granular LC domains with size down to 20 μm , much smaller than that (~ 200 μm) of ordinary nematic GO LCs ([Figure 2A](#)). In SEM inspections, the freeze-dried aerogels from GO FLCs demonstrate the ordered arrangement of GO sheets in much smaller regions ([Figure 2B](#)).

We further tracked the fragmentation process of GO LCs with increasing C_{KCl} ([Figures S6](#) and [S8](#)). As C_{KCl} increases, the macroscopically homogeneous nematic phase (≤ 5 mM) progressively breaks into finitized, locally ordered domains (7.5–20 mM) and eventually undergoes phase separation and aggregation into coagulations at higher ionic strengths (>20 mM). Further statistical results ([Figure S9](#)) show that with the decline of repulsion (increasing C_{KCl}), the LC domain size gradually decrease from nearly 200 μm down to about 20 μm ([Figure 2C](#)), comparable to the average size of GO sheets (15 μm ; [Figure S10](#)), indicating that the LC order can be frustrated down to single entity scale by screening repulsion. SEM images of freeze-dried LCs with a series of C_{KCl} also show that the alignment of GO sheets is gradually confined into small regions, indicating the fragmentation of LC domains ([Figure S6](#)). We additionally quantified the FLC domain size for GO samples with different lateral sizes. The FLC domain size increases monotonically with GO lateral size and remains comparable to it across all samples ([Figure S11](#)). This size coupling indicates a finite, sheet-governed orientational correlation length in the FLC state, providing direct support for

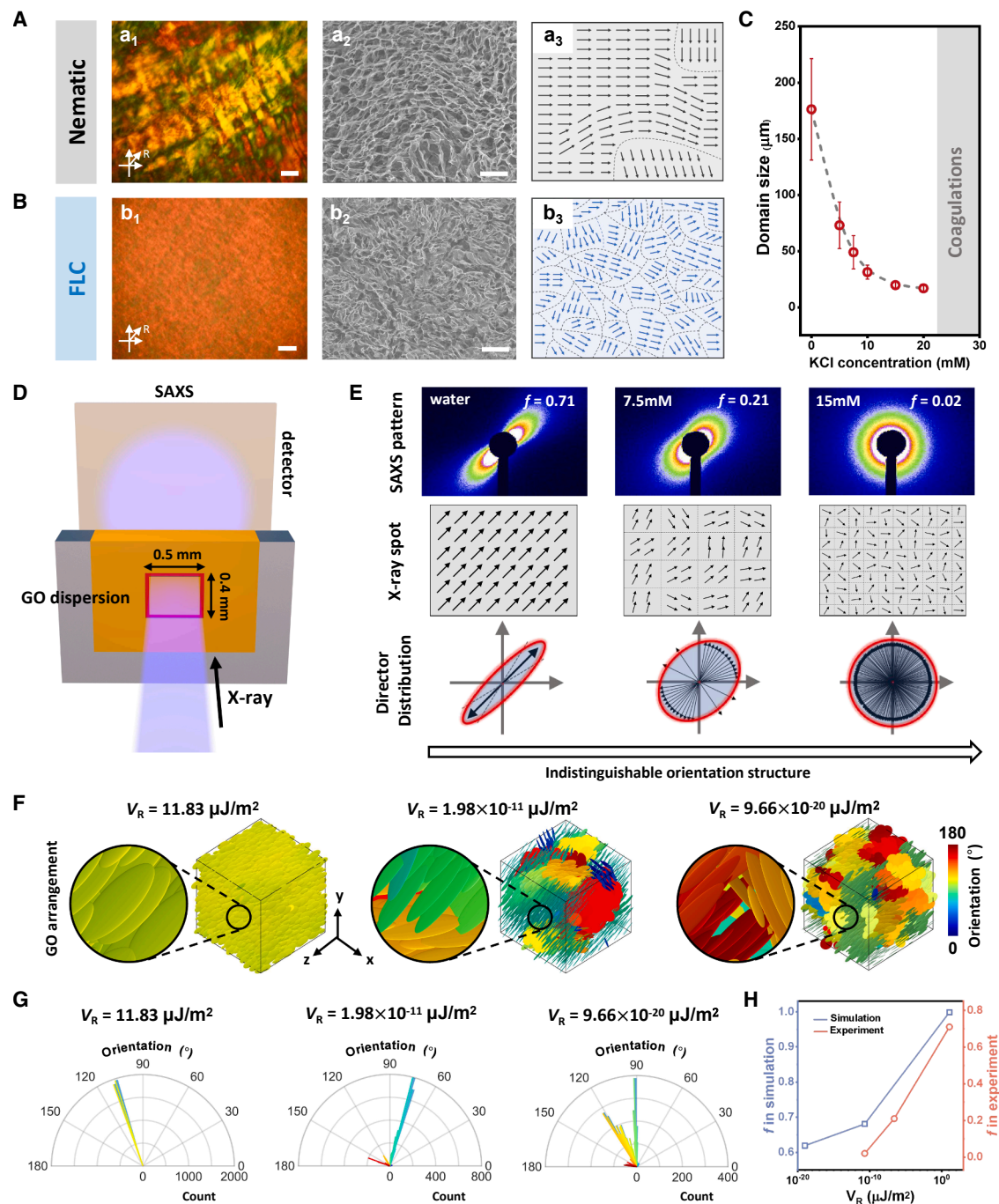


Figure 2. Structure analysis of GO FLCs and the LC fragmentation process

(A and B) The fragmented small LC regions in FLCs in POM (a_1 and b_1) and SEM images of freeze-dried GO dispersions (a_2 and b_2). The orientation of GO sheets in SEM images is shown in a_3 and b_3 . Scale bars: 200 μm in (a_1) and (b_1) and 20 μm in (a_2) and (b_2).

(C) The decreasing LC domain size with increasing C_{KCl} . Data are represented as mean $\pm$ SEM.

(D) Schematic illustration of SAXS tests of GO dispersions.

(E) The disappearance of orientation in SAXS patterns arising from the fragmentation of LC domains.

(F) The arrangement of GO in the CG-MD simulation. V_R denotes the repulsion energy density between GO sheets, which is calculated based on the DLVO theory (11.83 , 1.98×10^{-11} , and $9.66 \times 10^{-20} \mu\text{J}/\text{m}^2$ correspond to 0.1, 15, and 40 mM ion concentrations, respectively). The different orientation in the x-z plane is color-coded.

(G) The polar histogram quantifies the counts of orientations in the x-z plane.

(H) The same decreasing trend of order parameter f with V_R in experiments and MD simulations. The discrepancy in the f values is discussed in [Method S1](#).

the defining fragmentation of long-range nematic order. The nematic LC and FLC can reversibly switch by tuning the ionic strength, indicating that salt-induced FLC is a stable and reversible colloidal state rather than an irreversibly trapped aggregation intermediate (Figure S12). The time stability, ionic-strength reversibility, retained fluidity and sheet-size-dependent finite domain size collectively support the interpretation of salt-induced FLC as a stable intermediate LC state rather than a kinetically arrested transition toward aggregation (Note S2).

In small-angle X-ray scattering (SAXS) tests, we detected the fragmentation of LC orders during the transition to FLC (Figure 2D), in reminiscent of the spin order transition in ferromagnets.³⁴ As GO FLC forms, orientation order surprisingly disappears in SAXS patterns of FLCs, seemingly contradicting the presence of liquid crystallinity as observed in POM (Figure S13). In fact, this odd phenomenon is the very evidence of fragmentation of LC domains. In many condensed systems, such as LCs and ferromagnets, the macroscopic order does depend on the average summation of entities, like mesogens and spins.³⁴ In the SAXS tests, the size of X-ray spot is $0.4\text{ mm} \times 0.5\text{ mm}$, much larger than the FLC domains (down to $20\text{ }\mu\text{m}$). Similar to the dominating spin in ferromagnets transition as described by the Ising model, the fragmentation of LCs generates numerous small LC domains with random directors in the X-ray spots, which cancel each other out to exhibit fine powdery attributes. In this case, FLC does not exhibit dominating orientation in the macroscopic scale but has short distance orientation order as fragmentation means (Figure 2E).

We also tracked the rheology behaviors in the transition process from ordinary LC to FLC of GO (Figure S14). The viscosity, storage modulus (G'), and loss modulus (G'') all increase sharply as FLC gradually forms, because the motion of individual GO sheets in FLCs is hindered by neighboring LC domain with different orientations. As the steady FLC state forms, the viscosity decreases when C_{KCl} reaches 20 mM or higher due to the aggregation that tends to break into small pieces in flows. For the convenience and homogeneity, GO FLC with less than 15 mM KCl can be selected to suit materials processing. GO FLCs exhibit typical shear thinning behaviors as ordinary nematic GO LCs. The G' of GO FLCs far exceeds G'' in all frequency range, indicating the strong elasticity of this intermediate LC state, which can enable excellent plasticity and structure stability for fluid processing in spinning and 3D printing.

To validate the effect of electrostatic repulsion on the fragmentation behavior of GO LCs, we performed coarse-grained (CG) molecular dynamics (MD) simulations (Method S1; Figures S15 and S16; Table S1). To model the GO LCs, we adopted an ellipsoidal particle representation and calibrated the parameters of the Gay-Berne (GB) potential³⁵ based on the electrostatic repulsive potential derived from the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory.³⁶ Specifically, we analyze the evolution of the electrostatic repulsive potential energy between charged 2D colloids with respect to the colloid spacing at different ion concentrations based on the DLVO theory and the curves of three typical concentrations (0.1, 15, and 40 mM) are shown in Figure 3A. The surface repulsion energy densities (V_R) between colloids can be obtained from the values of the electrostatic repulsive potential energy curves at 80 nm, considering that

the equilibrium spacing between GO is typically estimated to be 80 nm.¹⁴ V_R at ion concentrations of 0.1, 15, and 40 mM are estimated as 11.83, 1.98×10^{-11} , and $9.66 \times 10^{-20}\text{ }\mu\text{J}/\text{m}^2$, respectively. In our simulations, we fitted the parameters of the GB potential based on the calculated electrostatic repulsion potential-spacing curves of the colloids, which are utilized to model the effect of V_R on the organization state of GO LCs. For systems with different surface repulsive energy densities, we analyzed the alignment and orientation (with respect to the x - z plane) (Figures 2F and 2G), and the nematic order parameter (f) of GO ($f = \langle 3\cos^2\theta - 1 \rangle / 2$, θ is the angle between GO basal plane and the selected reference orientation,¹ as shown in Figure 2H. GO sheets exhibit a uniform arrangement at large surface repulsive energy density ($V_R = 11.83\text{ }\mu\text{J}/\text{m}^2$). As the V_R decreases (e.g., $V_R = 1.98 \times 10^{-11}\text{ }\mu\text{J}/\text{m}^2$ and $V_R = 9.66 \times 10^{-20}\text{ }\mu\text{J}/\text{m}^2$), new disclination defects emerge to break the large orientation domain into multiple localized domains with random orientations. f decreases with decreasing V_R in both simulations and experiments, implying that the degree of fragmentation rises with reducing V_R (Figure 2H). In general, the simulation reflects the same trend of LC fragmentation as the experimental results, indicating the dominant role of reduced repulsion in this process. From the energy view, the strong repulsive interactions between GO sheets with larger V_R drive GOs to maintain large enough space and uniform arrangement to minimize the repulsive energy. In contrast, weaker repulsive interactions between GO sheets at lower V_R become insufficient to compensate the entropy loss of uniform arrangement and spontaneously drive the emergence of disclination defects to form FLC.

The interaction mechanism of GO FLCs

To elucidate the formation mechanism of FLC, we analyzed the intermolecular interaction in GO KCl system based on the DLVO theory.³⁶ The dispersion stability depends on the competition between the van der Waals attraction and electrostatic repulsion in charged colloidal systems (Figure 3A, upper image). The range of electrostatic repulsion can be characterized by the Debye screening length λ_D , which measures the thickness of the EDL. According to theoretical formulas and data,^{37,38} we calculated the pair potential of GO system as a function of C_{KCl} (Figure 3A, bottom image). The details of our calculation are listed in the Method S2 (Tables S2 and S3).

The reference inter-sheet distance (H) can be estimated as about 100 nm at 0.76 vol % GO dispersion based on the one-dimensional swollen model: $H = T/\Phi$, where T is the thickness of 2D sheets and is 0.8 nm for GO,¹⁵ Φ is the volume fraction.¹⁴ In this context, H is introduced only as an order-of-magnitude reference inter-sheet distance to facilitate comparison of how inter-sheet interactions evolve with ionic strength, rather than as a unique local structural spacing. The limitation and validity of this estimation is further discussed in Note S3. According to the above calculated interaction diagram, at low C_{KCl} down to 0.1 mM (where λ_D is about 31.3 nm), the strong long-range repulsion dominates even when H reaches 100 nm or larger, which prevents aggregation of GO sheets and drives the orderly alignment over long distance. The increase in ionic strength reduces the electrostatic repulsion by compressing the EDL and screening surface potential.³³ At high C_{KCl} (40 mM with λ_D of

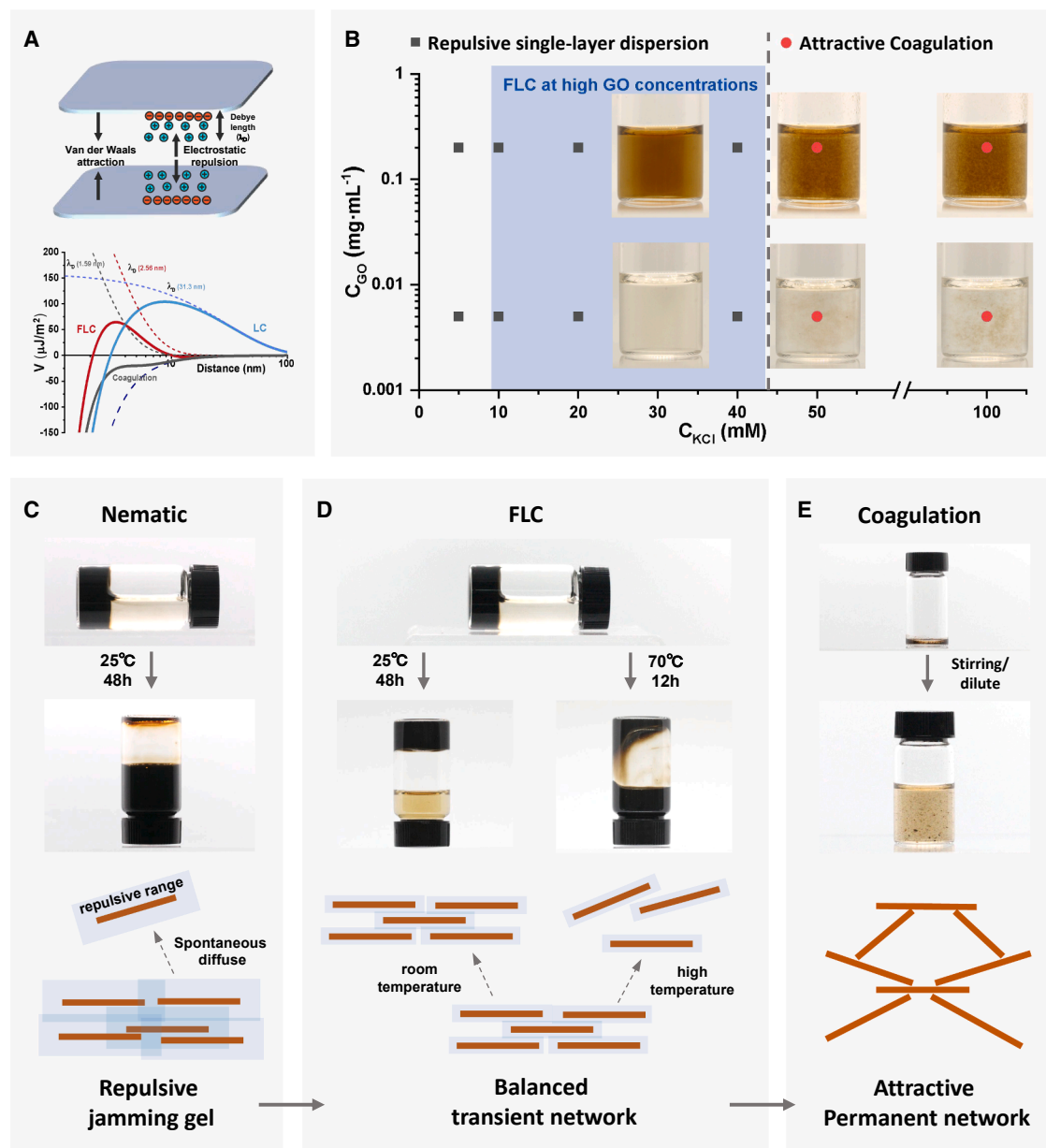


Figure 3. The interaction mechanism of GOFLCs

(A) Interactions between charged 2D colloids and the typical potential energy curves of nematic LC state (blue), FLC state (red), and coagulation state (gray). The long-dashed line represents van der Waals attraction potential energy.

Short-dashed lines represent the electrostatic repulsion potential energy under different ionic concentrations: 0.1 mM (blue), 15 mM (red), and 40 mM (gray). Solid lines are the total potential energy. The expressions of each potential energy curve are listed in Table S2.

(B) Dispersion states of dilute GO dispersions with different C_{KCl} , showing the dominated repulsion in GO FLCs.

(C) Dilution experiment of the repulsive jamming glass (also called repulsive gel) nature of nematic GO LCs.

(D) Dilution experiments of GO FLCs showing its transient networks. For both tests (C and D), dispersions of nematic GO LCs or GO FLCs at the same volume fraction (0.76 vol %) were carefully layered with pure water and maintained horizontally for an extended period. The final samples are shown inverted in the photographs only to highlight differences in their final states.

(E) Coagulated GO cannot redisperse after diluting for the formation of permanent attractive networks.

1.6 nm), attraction dominates over the entire distance range and GO sheets will irreversibly aggregate together by hydrogen bonding³⁹ and π - π stacking between the remaining conjugated mosaic domains of GO.⁴⁰

The pair potential at intermediate ionic concentrations (from 10 to 20 mM with λ_D between 3.2 and 2.2 nm) differs from the above two extreme cases, determining the different interaction feature of FLC. In this intermediate state, the interaction potential

is nearly flat at large distance (>10 nm), which suppresses long-range orientational coupling and drives the observed fragmentation of large LC domains. Meanwhile, at short distances of a few nanometers, well below the average distance between evenly dispersed GO sheets, repulsion still dominates, ensuring the single-layer dispersed state of GO sheets in FLC. This behavior is fundamentally distinct from irreversible aggregation (Figure 3B). Consistent with this interaction picture, GO dispersions remain homogeneous upon dilution at intermediate C_{KCl} (10–40 mM). In this regime, although the FLC state can be observed at high GO concentrations (Figure S18), dilute dispersions (<0.015 vol %) show no aggregation (Figures S19 and S20), contrary to the observed aggregates at higher C_{KCl} . Beyond this DLVO-based mesoscale picture, molecular-level solvent- and ion-mediated interfacial effects may also influence the interactions between GO sheets, especially near the LC-FLC-aggregation boundary.^{10,41,42}

Moreover, at near-contact separations, additional short-range interactions—such as hydrogen bonding and π - π stacking—can generate reversible inter-sheet contacts, giving rise to a transient network, as supported by rheological measurements (Figure S21). In strain-amplitude sweeps, GO FLCs exhibit a strong and robust low-strain elastic plateau and yield at much smaller deformation than large-area nematic LCs, consistent with stress transmission through abundant but reversible inter-sheet junctions. In contrast, nematic LCs show a weaker plateau and yield only at larger strains, reflecting mechanics dominated by reorganization of long-range orientational order rather than a connectivity-driven contact network.

The transient nature of the FLC network is further revealed by simple dilution tests that probe the diffusion of GO sheets from concentrated dispersions into water (Figures 3C–3E). GO dispersions of nematic LCs and FLCs with identical volume fractions (0.76 vol %) were carefully layered with water and then placed horizontally for an extended period. In ordinary nematic GO LCs, which behave as repulsive glass of the mutually jammed GO sheets (also called repulsive gel),^{43,44} strong electrostatic enables spontaneous diffusion of GO sheets into water at room temperature (Figure 3C). By contrast, GO FLCs show no noticeable diffusion at room temperature, reflecting reduced repulsion and the formation of transient inter-sheet network. Upon heating to 70°C, however, GO FLCs regain spontaneous diffusion (Figure 3D), indicating that the network is thermally reversible rather than permanent. This behavior is well supported reversible switching between G'-dominated (solid-like) and G''-dominated (liquid-like) responses in temperature step oscillatory rheology (Figure S21C). As to the extreme coagulation, attractive GO aggregations remain non-dispersible even when diluted to very low concentration by agitation (Figure 3E; Figure S22), indicating the interior permanent networks. The reversible transient network endows FLCs with favorable fluidity and processibility. Under uniaxial flowing, FLCs can show high orientation as ordinary LC behaves (Figure S23), suggesting the possible precise modulation of fragmented GO directors.

Heating-induced GO FLCs

We found that prolonged moderate heating (70°C) offers an alternative route to obtain GO FLCs. Similar to salt-induced FLC,

extended heating gradually reduced the LC domain size from ~ 200 to ~ 15 μm (Figures S24A, S24B, and S25), accompanied by the disappearance of orientation order in SAXS patterns (Figures S24C and S26) and a sharp increase in viscosity (Figure S24D). This thermally induced FLC also arises from the balance between attractive and repulsive interactions. Heating progressively removes oxygen-containing functional groups of GO, thereby decreasing surface charge density, as demonstrated by the declining C/O ratio in X-ray photoelectron spectroscopy (XPS; Figure S27C) and the continuous increase of zeta potential (Figure S27D). As a result, electrostatic repulsion between GO sheets gradually weakens during heating, eventually failing to sustain long-range order and leading to fragmentation of the LC domains. Due to irreversible chemical structural changes, the heating-initiated FLC cannot return back to the nematic LC phase after cooling (Figure S12). Further details are provided in Note S4.

Amorphous GO papers assembled from GO FLCs

For the good fluidic processibility, we applied GO FLCs to prepare prototype bucky papers by simple casting method and surprisingly found that GO FLCs generate a distinctive amorphous yet compact structure (Figure 4A), different from the polycrystalline GO laminate structure assembled from ordinary GO LCs (Figure S28). In our work, all LC-FLC paper comparisons are conducted at controlled thicknesses to eliminate thickness as a confounding factor. As direct evidences, the amorphous structure of GO FLC papers is demonstrated by X-ray diffraction (XRD) and transmission electron microscope (TEM) inspections. The cast GO FLC paper only has a nearly indistinguishable broad and weak diffraction peak in XRD profiles, in contrast with the sharp and strong diffraction peak of the polycrystalline paper derived from ordinary GO LCs (Figure 4B). In the XRD tracking, the intensity of the diffraction peak gradually decreases and the full width at half maximum (FWHM) stepwise increases from 0.56° to 2.08° as the formation of FLC with increasing C_{KCl} , corresponding to the greatly depressed packing in layering order (Figures S29A and S29B). Meanwhile, the peak shifts slightly to higher angles, corresponding to a reduced average interlayer spacing from 0.85 to 0.80 nm. We attribute this modest contraction to the smaller LC domains in the FLC state, which reduce large domain-boundary stacking defects during drying and enable denser packing with a smaller mean spacing.

High-resolution TEM inspection on the cast GO FLC paper cross-section (Figure 4C) also reveals the absence of long-distance crystalline order, even at a scale of 5 nm. As zooming into smaller scale, only few small layered order grains with width of ~ 1 nm are randomly distributed in the chaotic network, contrast to the long-distance order in GO papers cast by ordinary LCs (Figure S28). Moreover, SAXS and wide-angle X-ray scattering (WAXS) tests (Figures 4F and 4G) show that GO FLC papers have much lower Herman's orientation order (0.62 in SAXS tests and 0.73 in WAXS tests) than the control polycrystalline papers and other previously reported high-crystalline papers (about 0.9).^{19,45,46} Amorphous GO papers have also been prepared from the general FLC state triggered by other ions and thermal treatment (Figure S29C), indicating the intrinsic mechanism of FLCs for amorphous solid state.

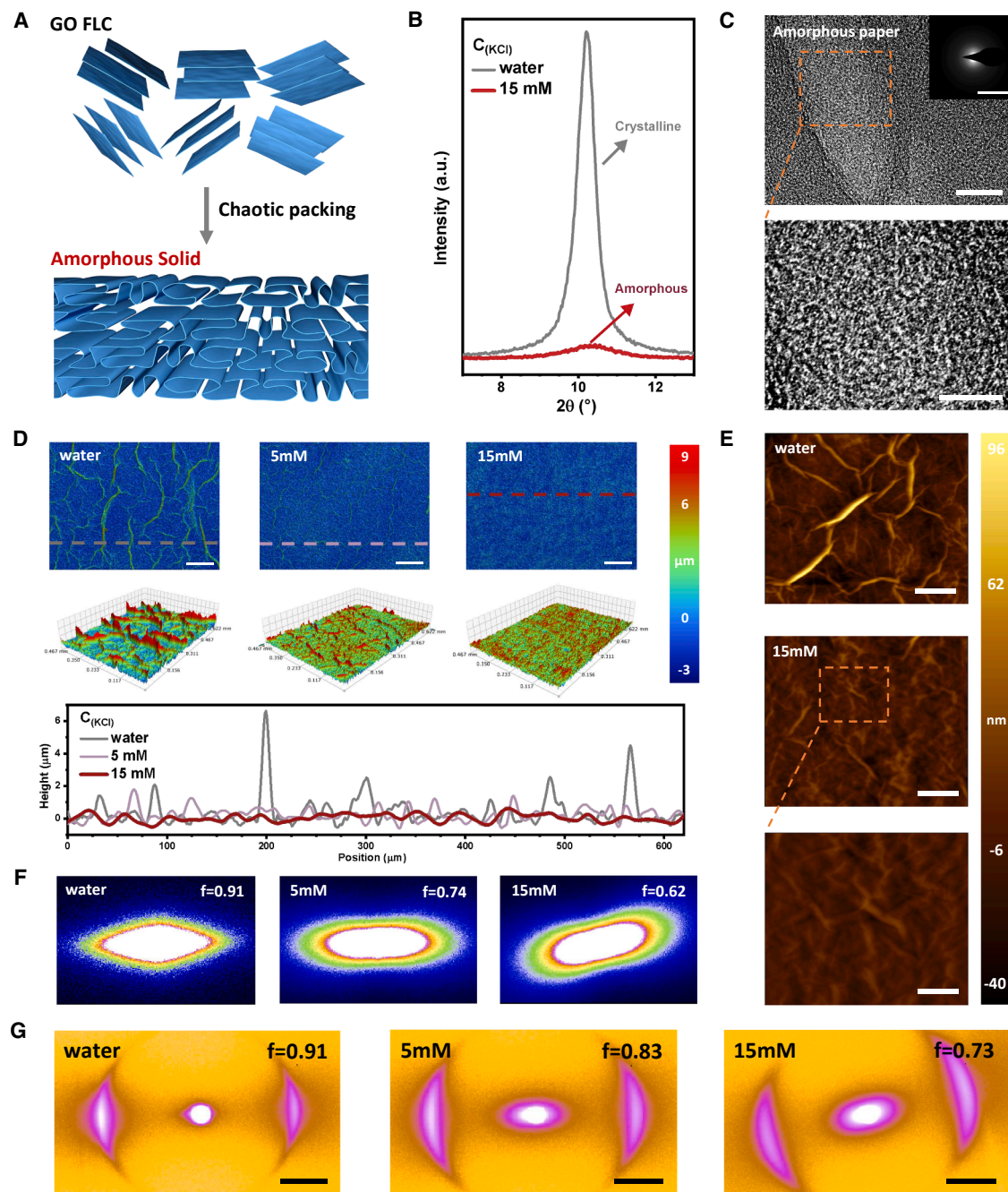


Figure 4. Amorphous attribute of GO FLC papers

(A) Schematic illustration of the assembly process from GO FLCs to amorphous GO solid.

(B and C) XRD profiles (B) and high-resolution TEM images and selected area electron diffraction (C) of GO FLC paper. Scale bars, 10 nm (upper image in C); 10 nm^{-1} (inset in the upper image); and 5 nm (lower image in C).

(D and E) Surface morphology of GO papers with increasing C_{KCl} measured by the white-light interferometer (D) and atomic force microscope (E).

Two-dimensional, three-dimensional images, and typical height distribution profiles are shown in (D). Scale bars, 100 μm (D); 2 μm (upper image in E); 1 μm (middle image in E); and 400 nm (bottom image in E).

(F and G) SAXS and WAXS patterns of GO papers with increasing C_{KCl} . Scale bars, 4 nm^{-1} (G).

We further tracked the formation mechanism of amorphous GO papers by multiscale structural analysis, directing an amorphous packing model of 2D sheets where their highly crumpled

conformation down to several nanometers hinders the ordered stacking. In the microscale, surface wrinkles become smaller and denser as the dispersions turn into FLCs (Figures S30–S32).

This wrinkle feature can be commonly found in GO FLC papers prepared by various methods (Figure S33). For the 10 μm thickness, polycrystalline GO papers have many high wrinkles above 2 μm , even up to 6 μm . The typical wrinkle height gradually decreases to about 0.5 μm as C_{KCl} increases from 0 to 15 mM, which corresponds to the decreasing alignment order from 0.91 to 0.62 as detected in SAXS tests (Figures 4D–4F; Figure S34). Closer AFM inspections at nanoscale show the same characteristics of wrinkles. Amorphous GO FLC paper features interwinding finer wrinkles with height of ~ 10 nm, decreasing from the large wrinkles (with height of ~ 80 nm) of ordinary GO papers (Figure 4E; Figure S35), which is also consistent with the decreasing order factor from 0.91 to 0.73 in WAXS tests (Figure 4G; Figure S36). The length (~ 500 nm) of fine wrinkles on amorphous paper is smaller than the average size of GO sheets, which denotes that individual GO sheets crumple to hinder the layered packing and generate the amorphous structure. Our structural analysis reveals that the flexibility of the assembly units is the key factor to achieve amorphous yet compact structure from the FLC state (Figure S37). For rigid entities, increased orientational disorder in the liquid phase typically leads to more packing defects and the formation of loosely packed structures. Alternatively, flexible entities can undergo crumpling or wrinkling to suppress layered stacking and fill defects arising from the orientation mismatch between LC domains, thereby promoting the formation of an amorphous and compact structure.

Wrinkle analysis indicates that fragmented domains in FLCs drive the formation of amorphous solids. During the assembly process, GO sheets typically stack as the solvent evaporates, producing ordered laminates accompanied by wrinkles induced by significant drying shrinkage at both the surface and interior.⁴⁷ This trend is evident in films cast from ordinary GO LCs, which display ordered lamellar structures with fewer but larger wrinkles (Figure 4D). In contrast, FLCs confine GO sheets within small domains comparable to the sheet size, which shrink locally without fusing together, as shown by SEM (Figure S38). Therefore, the confined GO sheets are only compressed locally but not merge in broad range to form large area laminates as in the case of ordinary LCs. In the deformation frame of thin films, larger and thicker deformation units tend to form bigger and fewer wrinkles.^{48,49} The localized shrinkage in range of GO sheets size generates smaller and denser wrinkles to disturb the ordered stacking and generate amorphous structure. As a comparison, moveable GO sheets in ordinary LC stack to generate thick skins on surface and larger laminates in interior, resulting in crystalline stacking with larger and fewer wrinkles.⁴⁷

Strong and tough amorphous papers

Amorphous GO papers processed from FLCs exhibit a cogeometric enhancement both in strength and toughness, outperforming polycrystalline papers from ordinary GO LCs (Figure 5A). The modulus, tensile strength, and toughness of 10- μm amorphous GO papers are tested as 6.42 ± 0.73 GPa, 434.8 ± 34.3 MPa, and 15.3 ± 1.5 MJ·m⁻³, which are 50%, 200%, and 500% higher than those of control polycrystalline papers respectively (Figure S39; Table S4). Amorphous reduced graphene papers exhibited the same enhancement trend with tensile strength up to 671.1 ± 23.4 MPa and toughness up to 18.3 ± 2.4 MJ·m⁻³.

The mechanical properties are continuously enhanced with increasing C_{KCl} , indicating that the simultaneous strengthening and toughening can be ascribed to the LC fragmentation process (Figure 5B), regardless of the trigger factors as ions or thermal treatments (Figure 5C). The amorphous structure of graphene laminates enables the simultaneous enhancement in strength and toughness, acting as an alternative option to promote the mechanical performance of graphene materials beyond the established crystallization.

Film thickness significantly affects the surface morphology and mechanical properties of GO papers (details in Note 5). With increasing thickness, FLC papers show larger surface height variations due to less uniform drying in thick films (Figure S47). Notably, this thickness-induced roughening differs from the severe, long-wavelength wrinkle networks characteristic of polycrystalline LC papers, which remain strongly suppressed in the FLC state even at high thickness. Although the mechanical performance of both LC- and FLC-derived papers decreases with increasing thickness from 5 to 20 μm , FLC papers consistently exhibit suppressed formation of large wrinkles and outperform LC papers at the same thickness at the same thickness (Figures S48 and S49; Tables S9 and S10), indicating an intrinsic advantage of the FLC assembly pathway rather than a thickness-driven artifact.

The improvement in mechanical properties can be attributed to the suppression of large, stress-concentrating flaws in the amorphous structure. This interpretation is consistent with Griffith-Irwin fracture mechanics, in which rare, large voids dominate crack initiation and catastrophic failure.⁵⁰ The surface and fracture morphologies provide direct support for this mechanism (Figures 4D and S41). In the polycrystalline films, the pronounced surface wrinkles are often associated with substantial internal cavities and can even be accompanied by interlayer delamination, as shown in the fracture images (Figures S41A and S41C). In contrast, the amorphous films exhibit fine, densely distributed wrinkles, consistent with more uniform densification of wrinkled GO sheets. Their fracture surfaces are correspondingly smooth and uniform, without failure features dominated by a single macroscopic flaw (Figures S41B, S41D, and S42). Further edge-profile analysis of independently prepared films confirms this trend, showing that the amorphous films have markedly lower RMS edge-height fluctuation and local fluctuation amplitude than the polycrystalline films, consistent with the reduced surface roughness and wrinkle height measured by white-light interferometry (Figures S32 and S43). This defect suppression is also reflected in the density. Despite lacking long-range ordered stacking, the amorphous papers show a higher density (2.01 g·cm⁻³) than the polycrystalline papers (1.79 g·cm⁻³; Figure S44; Table S5), indicating fewer large voids and a more compact load-bearing network. Finally, cross-sectional FIB-TEM provides microscopic evidence that polycrystalline papers contain large inter-domain stacking voids, whereas the amorphous papers form a more homogeneous, densely packed architecture of wrinkled sheets (Figure S45).

We further explored the effect of crumpled morphology on the mechanical properties of GOLC and GOFLC papers through a microstructure-aware theory. A deformable tension-shear (DTS) model is constructed, composed of staggered basic

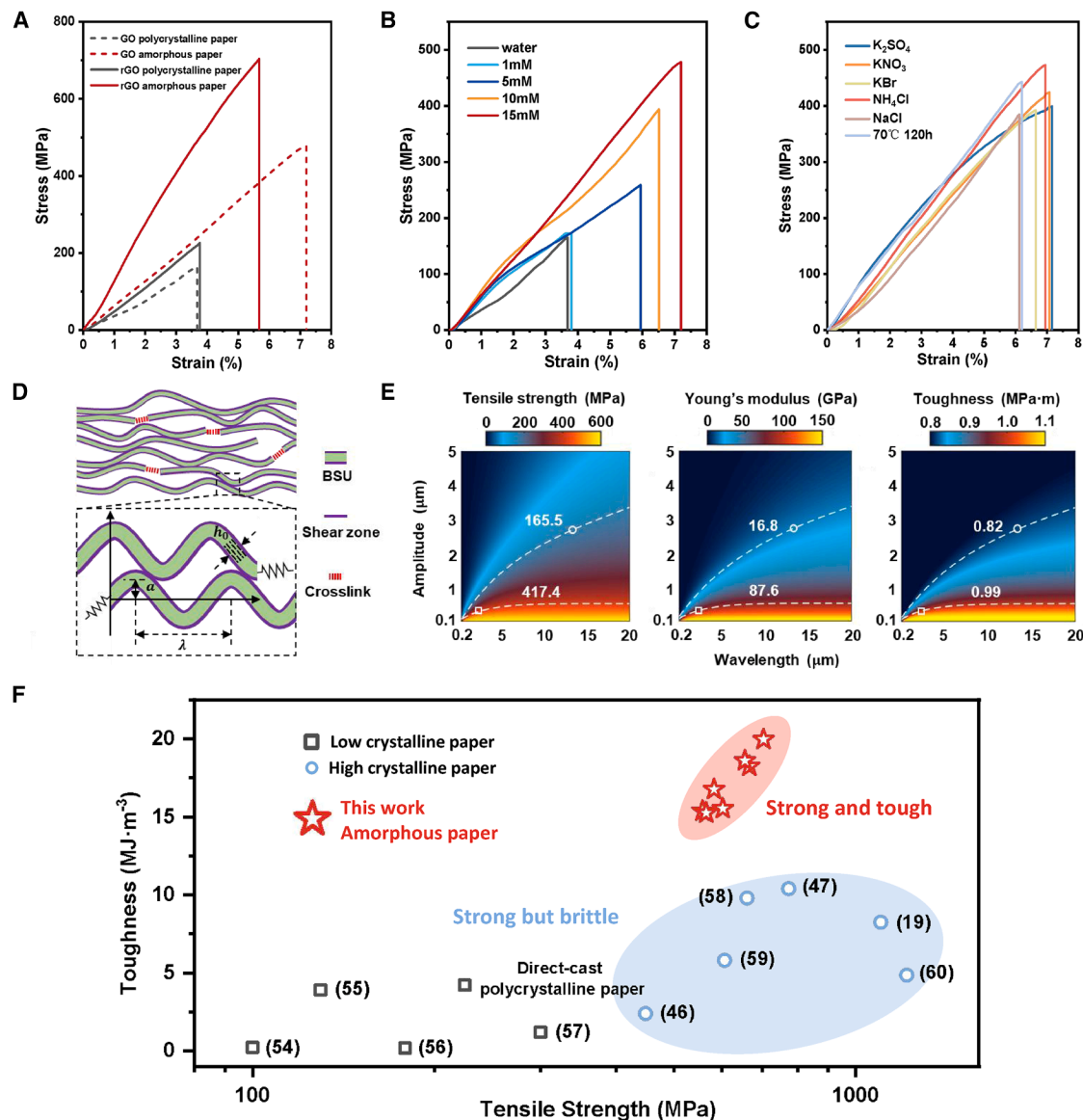


Figure 5. Mechanical properties of GO amorphous papers

(A) Typical stress-strain curves of GO polycrystalline and amorphous papers and corresponding reduced papers.

(B) The influence of KCl addition on the mechanical properties of GO papers.

(C) Mechanical properties of GO amorphous papers prepared by the addition of different monovalent salts or moderate heating.

(D) Schematic representation of the DTS model. The representative volume element (RVE) consists of staggered BSUs with morphological crumples, where the wavelength and amplitude of the crumples are λ and a , respectively, and the thickness of the BSU is h_0 .

(E) Calculated strength, modulus, and toughness in correlation with λ and a . White dashed lines show the contour profiles. Hollow circle and square denote large crumples ($\lambda = 13.4 \mu\text{m}$ and $a = 2.75 \mu\text{m}$) and small crumples ($\lambda = 2.5 \mu\text{m}$ and $a = 0.4 \mu\text{m}$), respectively, corresponding to experimental results in Figures S31 and S32.

(F) Comparison in strength and toughness of our amorphous rGO papers (Figure 5A; Figure S40) with previously reported high crystalline and low crystalline rGO papers (Table S6).

structural units (BSUs, closely packed GO laminates) in a layered arrangement (Figure 5D).⁵¹ Key mechanical properties are calculated as a function of wavelength and amplitude of the crumpled configuration (Figure 5E). The results show that, all the strength, modulus, and toughness increase with decreasing wavelength and amplitude, a more significant effect from the amplitude.

Wrinkles generate microscale cracks or voids (porosity also increases with misorientation) to disrupt the pathway of load transfer across the interfaces between the BSUs, resulting in reduced mechanical resistance to interface failure.^{51,52} Compared with polycrystalline GO LC papers, amorphous GO FLC papers exhibit finer wrinkles with smaller wavelengths and amplitudes.

Their lower porosity enables more efficient load transfer, leading to markedly enhanced mechanical properties.

The integration of both high strength (671 MPa) and toughness ($18.3 \text{ MJ}\cdot\text{m}^{-3}$) makes our amorphous graphene materials a distinctive material form, aside from the crystalline trend^{19,45,46,53–59} (Figure 5F). For neat graphene papers, improving the crystallinity of graphene stacking has generated high strength (up to 1.2 GPa) and stiffness.^{19,45,46,57–59} To conquer the unfavorable brittleness, multiple interlayer interactions have been proposed to relieve the conflict between strength and toughness.^{60–63} This proposed neat amorphous structure provides an alternative method to simultaneously promote the strength and toughness, using simple GO dispersion casting method and avoiding the complex design of compositions.

We further characterize the thermal and electrical conductivity of GO FLC papers using the laser flash analysis (LFA) and four-probe method (Figure S46; Tables S7 and S8). The reduced GO (rGO) FLC papers exhibit lower thermal conductivity ($\sim 34.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) and electrical conductivity ($\sim 3.19 \times 10^4 \text{ S}\cdot\text{m}^{-1}$) than the rGO LC papers ($\sim 39.4 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and $\sim 3.62 \times 10^4 \text{ S}\cdot\text{m}^{-1}$), consistent with the disrupted long-range alignment that enhances phonon and carrier scattering. Even so, the FLC-derived papers retain relatively high transport performance on an absolute scale while offering substantially improved mechanical robustness, highlighting a practical balance for applications requiring both reliability and efficient transport.

DISCUSSION

In summary, we identify FLC as an intermediate frustrated LC mesophase in 2D colloids between ordinary nematic LCs and coagulations. Unlike conventional nematic LCs, FLC state exhibits three distinct characteristics: intrinsic short-range cutoff of director correlations, balanced inter-sheet interactions, and transient inter-sheet network. Beyond its fundamental interest, the FLC state offers clear advantages for practical processing of 2D colloids. It can be accessed through simple and reversible ionic-strength modulation using common monovalent salts without specialized equipment, and is fully compatible with scalable solution-based techniques such as large-area blade coating (Figures S50–S52) and casting (Figure S53). Additional dip-coating tests on representative patterned substrates further demonstrate that GO FLCs can coat complex geometries with preserved surface patterns, with local thickness variations following a reproducible geometry-dependent trend (Figures S54 and S55; Note S6). By suppressing catastrophic defects, the FLC state can improve tolerance to processing-induced heterogeneities and enable more reliable and reproducible mechanical performance during large-scale processing. The resulting combination of high strength and toughness makes FLC-enabled materials attractive for mechanically demanding applications, such as flexible protective coatings, mechanically reliable barrier layers, and interfacial layers in composite systems. More broadly, the present findings suggest that analogous FLC states may be accessible in other highly anisotropic nano-sheet colloids when interparticle interactions and dispersion stability can be appropriately controlled. Establishing such generality in non-GO systems will require carefully optimized model

dispersions and remains an important direction for future work. For GO, the FLC state offers a distinctive processing route to accessing amorphous yet compact architectures of 2D sheets by deliberately disrupting long-range orientational order while preserving dense packing.

METHODS

Preparation of GO FLCs

Aqueous GO dispersion was purchased from Hangzhou Gaoxi Technology Co. Ltd (www.gaoxitech.com). The average size of the GO sheets was measured as $15 \mu\text{m}$ (Figure S10). The GO FLC dispersion can be prepared by simply mixing GO dispersions with monovalent salts aqueous solution of appropriate concentration such as KCl, NaCl, NH_4Cl , KBr, KNO_3 , K_2SO_4 , $(\text{NH}_4)_2\text{SO}_4$ aqueous solution, and so on. The volume fraction of GO (ϕ_v) is defined as:

$$\phi_v = \phi_m \left([1 - \phi_m] \frac{\rho_{\text{GO}}}{\rho_{\text{water}}} + \phi_m \right)^{-1},$$

where ϕ_m is the partial mass of GO in the dispersion, ρ_{water} is the density of water, and ρ_{GO} is the density of GO 1.32 g/mL .⁴⁴ A typical GO FLC dispersion was prepared by mixing 0.95 vol % GO dispersion and 75 mM KCl aqueous solution by the volume ratio of 4:1. For typical laboratory-scale preparations, representative total volumes were 5–10 mL, for example, by mixing 4 mL of GO dispersion with 1 mL of KCl solution or 8 mL of GO dispersion with 2 mL of KCl solution. The mixture was then placed at room temperature for 12 h to reach the equilibrium. The GO FLC dispersion can also be prepared by thermal annealing (70°C , more than 48 h) of GO dispersion (0.76 vol %) sealed in glass vessels under continuous stirring. To examine preparation-scale dependence, an $\sim 800 \text{ mL}$ FLC dispersion was prepared using the same formulation under continuous stirring.

Preparation of GO and rGO papers

Amorphous GO FLC papers with thickness of about $10 \mu\text{m}$ were prepared by cast drying on polyethylene terephthalate (PET) plates. The corresponding liquid layer of GO FLC (0.76 vol %) had a thickness of $\sim 1 \text{ mm}$ and was dried at 25°C (humidity $\sim 50\%$) for 24 h. The polycrystalline GO papers were prepared by the same procedure as GO FLC papers, except no salt addition or heating. All rGO papers were prepared by chemical reduction with a mixed ethanol/aqueous 47% hydroiodic acid solution (3/1 by volume) at 90°C for 12 h. Then, the resultant rGO papers were repeatedly washed by ethanol to remove residual iodine and dried at room temperature for 12 h. Film thickness was tuned during blade coating by adjusting the GO concentration and wet-film thickness, and the final thickness of each freestanding film was determined by cross-sectional SEM and micrometer.

Preparation of GO coatings

GO coatings were prepared by a dip-coating process. Briefly, the target regions of objects were immersed in either the ordinary GO LC or GO FLC dispersion and held still for 5 s to wet the surface. The objects were then slowly withdrawn from the dispersion and kept vertically for 30–60 s to allow excess dispersion

to drain off. The coated objects were subsequently dried at room temperature in a partially covered container without forced airflow to allow gradual solvent evaporation and to reduce drying-induced defects. The thickness of the coatings is affected by the concentration of the dispersion, the viscosity of the dispersion and the number of dip coatings. Typically, we used the 0.6 vol % GO dispersion with the addition of 15 mM KCl for GO FLC coatings on simple surfaces and the 0.3 vol % GO dispersion with the addition of 40 mM KCl for GO FLC coatings on complex curved surfaces.

Characterizations

Mechanical testing was conducted using an XQ-2 instrument (Shanghai New Fiber Instrument Co., Ltd.) at a loading rate of 2 mm·min⁻¹ with a gauge length of 5 mm. GO/rGO papers (~10 mm × 1 mm) were fixed onto a 5 × 5 mm paper frame using epoxy resin. After curing, the frame was mounted on the XQ-2 fixtures, and both sides of the frame were carefully cut to isolate the sample for testing. Thickness was measured three times using a micrometer, and the average value was used. Width was determined via optical microscopy, and the cross-sectional area was calculated as the product of thickness and width. Mechanical properties were obtained by averaging at least five representative stress-strain curves. SEM images were taken on Hitachi S4800 field emission system. For SEM characterization of LC structure, thin liquid films were rapidly frozen by liquid nitrogen to inhibit the growth of large ice crystals that would disrupt sheet alignment (Figure S7). POM and OM observations were performed on a Nikon E600POL. AFM images were collected by Cypher ES Environmental AFM. Zeta potential tests were taken on a Malvern Zetasizer Nano ZS90. Rheological tests were taken on the Discovery HR-10 of TA instrument. The white light interference microscopy images were collected on Veeco NT 9100. XRD profiles were collected on a LabX XRD-6000 using monochromatic Cu 17 K_{α1} radiation ($\lambda = 1.5406 \text{ \AA}$) at 40 kV. Synchronous SAXS tests of GO dispersions were carried out in BL16B1 beamline station of Shanghai Synchrotron Radiation Facility (SSRF). GO dispersions were directly injected into the sample cell using a syringe and gently shaken to ensure complete filling. Throughout the process, stirring was avoided to prevent disruption of the initial LC domains. SAXS and WAXS were taken on XEUS 3.0 SAXS/WAXS System. All data were collected by subtracting the background scattering of air. The crystalline microstructure was characterized by transmission electron microscope (FEI Tecnai G2 F20 S-TWIN) and the TEM samples were prepared by ion-milling (FEI Quanta3D FEG). The thermal transport properties were characterized by LFA, where the in-plane thermal diffusivity was measured by LFA 467 of NETZSCH, the density was obtained from the solvent suspension method and the specific heat capacity was measured by DSC 214 of NETZSCH. The electrical conductivity was characterized by the four-probe method using the Keithley 2460 Source Measure Unit (SMU).

Calculation based on the DLVO theory

The Debye length $\lambda_D = \kappa^{-1}$ was calculated by the following equation:

$$\kappa = \sqrt{10Cz^2} [\text{nm}^{-1}], \quad (\text{Equation 1})$$

where C is the concentration of ions, and z is the number of charges each ion has. In our study, the Debye length of 0.1, 15, and 40 mM GO dispersions can be calculated as 31.3, 2.7, and 1.6 nm, respectively.

The dispersion stability depends on the competition between the van der Waals attraction and electrostatic repulsion in charged colloidal systems. The van der Waals attraction potential energy V_A between two colloidal plates was calculated by

$$V_A = -\frac{A}{12\pi H^2} [\text{J/m}^2], \quad (\text{Equation 2})$$

where A is the Hamaker constant and H is the distance between the plates. The Hamaker constant of GO can be referred as $4.9 \times 10^{-20} \text{ J}$.³⁷

The electrostatic repulsion potential energy was calculated by the following equation:

$$V_R = 2\epsilon\kappa\phi_d^2 e^{-\kappa H} [\text{J/m}^2], \quad (\text{Equation 3})$$

where ϕ_d can be obtained from zeta potential measurements (Figure S17). The total potential energy was obtained by adding up the van der Waals attraction potential energy V_A and the electrostatic repulsion potential energy V_R . More details about the DLVO theory can be found in the supplemental method 2.

Calculation of order parameters

The orientation of SAXS and WAXS patterns was quantified by converting the orientation distribution to a Herman's order parameter, f , defined as

$$f = \int_0^\pi I(\varphi) \left(\frac{3}{2} \cos^2 \varphi - \frac{1}{2} \right) \sin(\varphi) d\varphi. \quad (\text{Equation 8})$$

where φ is the azimuthal distribution and $I(\varphi)$ corresponds the density at φ . The intensity is normalized according to

$$\int_0^\pi I(\varphi) \sin(\varphi) d\varphi = 1. \quad (\text{Equation 9})$$

SAXS was used to characterize the orientation order on tens of nanometer scale. WAXS was applied to evaluate the order parameters at the atomic scale.⁴⁷

Microstructure-aware theoretical modeling

Microstructure-aware theories such as the shear-lag model⁵¹ and the DTS model⁶⁴ can address the effect of microstructural complexity on the mechanical properties of macroscopic assemblies of GO, including morphological crumples, polydispersity, as well as their intra-layer and inter-layer interactions. In this study, the DTS model is employed to analyze the mechanical responses of GO LC and GO FLC papers, which consist of multiple crumpled BSUs (or closely packed laminates), as shown in Figure 5D. The details of constructing microstructure-aware theories can be found in the supplemental method 3 and our previous work.⁵¹ Key mechanical properties including effective Young's modulus, tensile strength, and toughness are calculated as a function of wavelength and amplitude of the crumpled configuration based on

this DTS model. Here, the material parameters used for the theoretical analysis of the mechanical response of the GO LC and GO FLC papers are as follows^{65–68}: $Y = 250$ GPa, $G = 25$ GPa, $\gamma_{cr} = 0.25$, $h_0 = 0.01$ μm , $l = 20$ μm , and $\omega = 2$. The value of k_e is set to 0 to ignore the crosslinking of BSUs in the same layer so that the interface between BSUs dominates the load-bearing capacity. The amplitude a and wavelength λ of the crumpled morphology of the BSUs are considered in the range of $0.1\ \mu\text{m} \leq a \leq 5\ \mu\text{m}$ and $0.2\ \mu\text{m} \leq \lambda \leq 20\ \mu\text{m}$, respectively, which cover the amplitude and wavelength of the experimentally observable crumples.

RESOURCE AVAILABILITY

Lead contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the lead contact, Zhen Xu (zhenxu@zju.edu.cn).

Materials availability

The methods sections in the main text and [supplemental information](#) include material sources in the study, and further information is available from the [lead contact](#) upon reasonable request.

Data and code availability

All the necessary data have been provided in the main text and [supplemental information](#). Any additional data related to this paper can be obtained from the [lead contact](#) upon request. This study did not generate/analyze datasets or code.

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AUTHOR CONTRIBUTIONS

Z.X., C.W.S., Y.G., and S.P.L. conceived the idea; C.W.S., Y.C.L., Z.X., and Z.P.X. designed the experiment; C.W.S. and C.D.G. set up the experimental equipment; C.W.S., X.Y., and G.F.C. contributed to the preparation of materials; C.W.S. and S.P.L. carried out tests; all authors analyzed experimental data; Y.C.L. and Z.P.X. designed and performed theoretical modeling; C.W.S. and Y.C.L. led the writing and modification of the paper; Z.X., Z.P.X., Y.J.L., P.L., and C.G. supervised the work.

DECLARATION OF INTERESTS

The authors declare no competing interests.

SUPPLEMENTAL INFORMATION

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