

Rheology of Cubic Blue Phases

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We study the behaviour of cubic blue phases under shear flow via lattice Boltzmann simulations. We focus on the two experimentally observed phases, Blue Phase I (BPI) and Blue Phase II (BPII). The disclination network of Blue Phase II continuously breaks and reforms under steady shear, leading to an oscillatory stress response in time. For larger shear rates, the structure breaks up into a Grandjean texture with a cholesteric helix lying along the flow gradient direction according to the imposed flow. BPI leads to a very different response. Here, oscillations are only possible for intermediate shear rates – very slow flow leads to a transition into an amorphous network with an apparent yield stress. Larger shear rates can make the network sensitive to flow-induced transitions to metastable and amorphous states. For even larger flow rates a Grandjean texture is observed, just as in the case of BPII. At the highest flow rates both cubic blue phases adopt a flow-aligned nematic state. Our results provide the first theoretical investigation of sheared blue phases in large systems, and are relevant to understanding the bulk rheology of these materials.

1 Introduction

Cholesterics are liquid crystals in which the local nematic director field shows spontaneous twist in thermodynamic equilibrium¹. The simplest manifestation is the standard cholesteric phase where the director precesses around a single helical axis of fixed orientation. For highly chiral systems, however, the preferred configuration close to the isotropic boundary features twist around two perpendicular axes, as opposed to just one axis in the regular cholesteric state, and the corresponding deformation is denoted a “double-twist cylinder”. As it is topologically impossible to cover continuously 3D space with double-twist cylinders, defects arise. The resulting disclination lines (at which the nematic director is undefined) organise into a variety of regular periodic lattices, giving rise to the so-called cubic blue phases (BPs)^{2,3}. There are two experimentally observed cubic blue phases, BPI and BPII (a third, BPIII, is thought to be amorphous⁴).

BPs were long considered as purely of academic interest due to their very narrow range of stability. This view has changed since the creation of polymer-stabilised and other thermally stabilised BPs^{5,6}, which has opened up the possibility of novel applications. During the last few years considerable progress has been achieved regarding the behaviour of BPs in confined geometries^{7–9}, under external fields^{10–14},

and in the presence of colloidal particles¹⁵. The kinetics of BP domain growth have been recently addressed¹⁶. However, our understanding of their dynamical behaviour under flow remains very limited. The aim of this work is to address this issue by studying, for the first time, the response of large BP samples to a shear flow.

Flow response in cholesterics is both strongly non-Newtonian and highly anisotropic. For example, if a standard cholesteric phase is subjected to a Poiseuille flow along its helical axis, small pressure differences drive flow mainly through “permeation”, as first investigated by Helfrich¹⁷. In the permeation mode the liquid crystal flows while leaving the director field virtually unchanged, which leads to high dissipation and large viscosities. Marenduzzo et al.^{19,20} simulated shear and Poiseuille flow in cholesteric liquid crystals in the permeation mode, and showed the importance of the boundary conditions in determining the apparent viscosity of the fluid. They also found that a strong secondary flow appears. Early experiments with cholesteric liquid crystals showed that flow can also give rise to conformational transitions.¹⁸ Rey^{21,22} studied shear in cholesterics oriented with the helix along the vorticity axis and found that, at low Ericksen number, travelling twist waves appear which lead to the rotation of the cholesteric helix. At higher forcing, the helix uncoils, creating a flow-induced nematic phase. These result, however, were derived under the assumption that the molecules rotate only in flow-gradient plane whilst the orientation of the cholesteric helix remained unchanged. Rey also studied cholesterics subjected to both steady flow and low frequency small amplitude oscillatory shear for different helix orientations^{23,24}. He found that splay/bend/twist deformations were excited when

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the helix was aligned along the flow direction; splay/bend deformation occurred when the helix was aligned along the velocity gradient; but only twist deformations appeared when the helix was aligned along the vorticity axis.

Dupuis et al.²⁵ performed the first numerical investigation of BP rheology in Poiseuille flow, starting from equilibrium structures of BPI and BPII and a periodic array of doubly twisted cylinders. Under small forcing, the network opposed the flow, giving rise to a significant increase in apparent viscosity. Upon increasing the forcing they found clear evidence of shear thinning. In the crossover region they predicted a novel oscillatory regime where the network continuously breaks and reforms as portions of the disclinations in the centre of the channel move to neighbouring cells and re-link with the parts of the network left behind by the flow. The viscosity still decreases with forcing (the system shear thins) but much less than for cholesterics in the permeation mode, which is in agreement with experiments^{26,27}.

Our work differs from these earlier efforts as it addresses homogeneous shear flows, and focuses on flow-induced reconstruction and nonequilibrium transition between different blue phase networks, which appear at intermediate or high shear. Our simulations employ Lees-Edwards boundary conditions, which are naturally suited to address these regimes, and bulk as opposed to boundary-dominated flow. Our simulations are large scale and parallel, so that we can study significantly larger systems than previously possible, and our results can in principle be compared with bulk rheology experiments.

Our paper is organised as follows. In Section II, we describe the method we use and review the hydrodynamic equations of motion which we aim to solve. In Section III, we report our numerical results, separating them into subsections referring to Blue Phase I and Blue Phase II, and corresponding to low, intermediate, and high shear. Finally, we draw our conclusions in Section IV.

2 Model and Methods

Our approach is based on the well-established Beris-Edwards model for hydrodynamics of cholesteric liquid crystals²⁸, which describes the ordered state in terms of a traceless, symmetric tensor order parameter $\mathbf{Q}(\mathbf{r})$. In the uniaxial approximation, the order parameter is given by $Q_{\alpha\beta} = q_s(\hat{n}_\alpha \hat{n}_\beta - \frac{1}{3}\delta_{\alpha\beta})$ with $\hat{\mathbf{n}}$ the director field and q_s the amplitude of nematic order. More generally, the largest eigenvalue of \mathbf{Q} , $0 \leq q_s \leq \frac{2}{3}$ characterises the local degree of orientational order. The thermodynamic properties of the liquid crystal are determined by a free energy \mathcal{F} , whose density f consists of a

bulk contribution f_b and a gradient part f_g , as follows,

$$\begin{aligned} f_b &= \frac{A_0}{2} \left(1 - \frac{\gamma}{3}\right) Q_{\alpha\beta}^2 \\ &\quad - \frac{A_0\gamma}{3} Q_{\alpha\beta} Q_{\beta\gamma} Q_{\gamma\alpha} + \frac{A_0\gamma}{4} (Q_{\alpha\beta}^2)^2, \\ f_g &= \frac{K}{2} (\varepsilon_{\alpha\gamma\delta} \partial_\gamma Q_{\delta\beta} + 2q_0 Q_{\alpha\beta})^2 + \frac{K}{2} (\partial_\beta Q_{\alpha\beta})^2. \end{aligned} \quad (1)$$

The first term contains a bulk-free energy constant A_0 and the temperature-related parameter γ which controls the magnitude of order. The second part quantifies the cost of elastic distortions, which is proportional to the elastic constant K ; we work for simplicity in the one-elastic constant approximation¹. The wavevector q_0 is equal to $2\pi/p_0$, where p_0 is the cholesteric pitch. The actual periodicity of the BP structure, p , does not need to be equal to p_0 . Indeed, the “redshift” $r = p/p_0$ is adjusted during the equilibration phase of the simulation, to optimise the free energy density before shearing begins – this is done by following the procedure previously described in²⁹.

A thermodynamic state is specified by two dimensionless quantities: the reduced temperature

$$\tau = \frac{27(1 - \gamma/3)}{\gamma}, \quad (2)$$

which vanishes at the spinodal point of a nematic ($q_0 = 0$), and the reduced chirality

$$\kappa = \sqrt{\frac{108Kq_0^2}{A_0\gamma}}, \quad (3)$$

which measures the ratio of gradient to bulk free energy.

The dynamical evolution of the order parameter is given by the equation

$$(\partial_t + v_\alpha \partial_\alpha) \mathbf{Q} - \mathbf{S}(\mathbf{W}, \mathbf{Q}) = \Gamma \mathbf{H}. \quad (4)$$

The first term on the left hand side of Eq.4 is a material derivative, which describes the rate of change of a quantity advected by the flow. The second term accounts for the rate of change due to local velocity gradients $W_{\alpha\beta} = \partial_\beta v_\alpha$, and is explicitly given by²⁸

$$\begin{aligned} \mathbf{S}(\mathbf{W}, \mathbf{Q}) &= (\xi \mathbf{A} + \Omega)(\mathbf{Q} + \frac{\mathbf{I}}{3}) \\ &\quad + (\mathbf{Q} + \frac{\mathbf{I}}{3})(\xi \mathbf{A} - \Omega) - 2\xi(\mathbf{Q} + \frac{\mathbf{I}}{3}) \text{Tr}(\mathbf{QW}), \end{aligned} \quad (5)$$

where Tr denotes the tensorial trace, while $\mathbf{A} = (\mathbf{W} + \mathbf{W}^T)/2$ and $\Omega = (\mathbf{W} - \mathbf{W}^T)/2$ are the symmetric and antisymmetric part of the velocity gradient, respectively. ξ is a constant depending on the molecular details of the liquid crystal. Flow alignment occurs if $\xi \cos 2\theta = (3q_s)/(2 + q_s)$ has a real solution for θ , the so-called Leslie angle: we select this case by

setting $\xi = 0.7$ in our simulations. \mathbf{H} is the molecular field, which is a functional derivative of \mathcal{F} that respects the tracelessness of \mathbf{Q} :

$$\mathbf{H} = -\frac{\delta \mathcal{F}}{\delta \mathbf{Q}} + \frac{\mathbf{I}}{3} \text{Tr} \left(\frac{\delta \mathcal{F}}{\delta \mathbf{Q}} \right). \quad (6)$$

The rotational diffusion constant Γ in Eq. 4 is proportional to the inverse of the rotational viscosity $\gamma_1 = 2q_s^2/\Gamma^1$.

The time evolution of the fluid density and velocity are respectively governed by the continuity equation $\partial_t \rho = -\partial_\alpha(\rho v_\alpha)$, and the following Navier-Stokes equation:

$$\partial_t v_\alpha + \rho v_\beta \partial_\beta v_\alpha = \partial_\beta \Pi_{\alpha\beta} + \eta \partial_\beta [\partial_\alpha v_\beta + \partial_\beta v_\alpha]. \quad (7)$$

This emerges from the Chapman-Enskog expansion of the lattice Boltzmann (LB) equations that we solve numerically. A further term $\eta(1+3\frac{\partial P_0}{\partial \rho})\partial_\mu v_\mu \delta_{\alpha\beta}$ that formally appears in this expansion is negligible under the slow flows considered here for which the the fluid motion is almost incompressible³⁰. η is an isotropic background viscosity which is set to $\eta = 0.6666$ in LB units (these are discussed below, see^{4,16}). The thermodynamic stress tensor reads explicitly

$$\begin{aligned} \Pi_{\alpha\beta} &= P_0 \delta_{\alpha\beta} - \xi H_{\alpha\gamma} \left(Q_{\gamma\beta} + \frac{1}{3} \delta_{\gamma\beta} \right) \\ &- \xi \left(Q_{\alpha\gamma} + \frac{1}{3} \delta_{\alpha\gamma} \right) H_{\gamma\beta} + Q_{\alpha\gamma} H_{\gamma\beta} - H_{\alpha\gamma} Q_{\gamma\beta} \\ &+ 2\xi \left(Q_{\alpha\beta} + \frac{1}{3} \delta_{\alpha\beta} \right) Q_{\gamma\nu} H_{\gamma\nu} - \partial_\alpha Q_{\gamma\nu} \frac{\delta \mathcal{F}}{\delta \partial_\beta Q_{\gamma\nu}} \end{aligned} \quad (8)$$

and is responsible for strong non-Newtonian flow effects. In the isotropic state $\mathbf{Q} \equiv 0$ and Eq.8 reduces to the scalar pressure as would appear in Eq. 7 for a Newtonian fluid.

We next define a dimensionless number that describes the deformation of the director field under flow. This so-called Erickson number is given by

$$Er = \frac{\eta v l}{K} \quad (9)$$

with η and K defined previously, and v and l a typical velocity and length scale. In the present work $l = p_0/2 = \pi/q_0$ was used as this is the approximate size of the BP unit cell. Likewise, v was taken to be the velocity difference across one unit cell, i.e. $v = \gamma l$. **The elastic constants were $K = 0.02$ (BPII) and $K = 0.04$ (BPI), respectively.**

The system of coupled partial differential equations 4 and 7 is solved by means of a hybrid method³¹ which uses a combination of lattice Boltzmann and finite difference schemes. (This is in contrast with some earlier methods using solely LB^{30,32}.) The Navier-Stokes equation is solved via the lattice Boltzmann approach, using a standard three-dimensional

model with 19 discrete velocities (D3Q19). A regular lattice with spacing $\Delta x = \Delta y = \Delta z = 1$ is used and the time step is $\Delta t = 1$ in lattice units. Coupling to the thermodynamic sector is via a local body force computed as the divergence of the thermodynamic stress Eq. 8; the resulting velocity field is used in the computation of the time evolution of \mathbf{Q} via a standard finite difference method using the same grid and the same time step as the LB. The system is periodic in all three coordinate directions. Sliding boundary conditions, or Lees-Edwards planes, are explicitly implemented in both hydrodynamic and thermodynamic sectors to impose shear on the system. On crossing a Lees-Edwards boundary a Galilean transformation is applied with a velocity increment which is fixed in time (and limited in size by the low Mach number constraint of LB). An appropriate transformation of LB distributions which propagate across a boundary³³ must be applied at each time step, and appropriate adjustment to $W_{\alpha\beta}$ is required to compute cross-plane gradients of the velocity field used in the update to \mathbf{Q} . In both cases interpolation of the relevant quantities is required to cope with the relative displacement of neighbouring lattice sites separated by a sliding plane (the displacement may be a fraction of a lattice unit at any given time step). The use of multiple sliding planes equally spaced in a single system allows the overall shear rate to be maintained indefinitely as the system becomes larger in the velocity gradient direction (in contrast with the use of solid walls to impose shear). This hybrid method with Lees-Edwards planes has been used successfully to study e.g., smectics³⁴.

In the following we report results of simulations of the bulk flow behaviour of the cubic blue phases BPI and BPII. Typical runtimes for system size $L_x \times L_y \times L_z = 128^3$ on 512 processes were in the region of 18 to 24 hours. The timestep and lattice spacing in lattice Boltzmann units (LBU) can be mapped approximately to $\sim 1\text{ns}$ and $\sim 10\text{nm}$ in SI units, respectively. The LB unit of stress is equal to about 10^8 Pa . Further details about the conversion from LBU to SI units can be found elsewhere^{4,16}. In what follows, x , y and z denote respectively the velocity, velocity gradient and vorticity direction; Π_{xy} is therefore the shear stress.

3 Results and Discussion

For typical simulations reported in this work, we chose as initial conditions thermodynamic states that are well inside the equilibrium region of the individual blue phase, and far away from the cholesteric-isotropic transition. Thus, temperature and chirality were $\tau = -0.5$, $\kappa = 1.0$ in case of BPI and $\tau = -0.5$, $\kappa = 2.0$ for BPII, respectively. For these parameters the total free energy density f remained always negative at all flow rates simulated. Since by Eq. 1 $f = 0$ for an isotropic phase with $\mathbf{Q} \equiv 0$, this means that our system, which is never far from equilibrium locally even under flow, always remains

in a liquid crystalline state. We also performed selected simulations on metastable states at higher and lower temperatures, and at different chiralities, but did not find any significant differences in the general flow behaviour from that described below. The only quantitative difference we found was that, for thermodynamic states that are closer to the phase boundary than the one we focused on (and describe below), the critical shear rate at which the disclination network broke up into chiral nematic states was lower. This is expected as these states have on average higher free energy densities and smaller order parameters than those we focus on in what follows, and cannot therefore withstand the same external forces before breaking down.

As usual in BP simulation studies^{4,16}, we initialised our runs with analytical solutions that minimise the free energy functional Eq. 1 in the high-chirality limit and equilibrated these configuration for 5000 LB timesteps before we started the shear flow. During the equilibration sequence the optimal redshift r was calculated and applied at every timestep. After equilibration the redshift was kept constant throughout the rest of the simulation with shear flow (as it is no longer justified to optimise the free energy in a nonequilibrium scenario). We chose a pitch length of 32 LBU for BPII and 64 LBU in case of BPI, and we considered in both cases 4 unit cells along each coordinate direction, for a total of 64 unit cells in our simulation box. Runs with higher resolution confirmed that this choice was adequate to track all kinematic details of the blue phase networks in shear flow. This includes reconstruction of the unit cell not accessible in simulation with only few cells²⁵.

Simple shear flow was imposed by means of the Lees-Edwards boundary conditions with the top (bottom) part of the system flowing in the positive (negative) x -direction and the velocity gradient along the y -direction. The shear rates were varied over more than two orders of magnitude from about $\dot{\gamma} = 2.44 \times 10^{-6}$ to 1.875×10^{-3} LBU. For clarity we classify various flow regimes, namely three in the case of BPII and five in the case of BPI. These regimes comprise those with periodically recurring conformations and oscillatory response (PRC), amorphous networks (AN), amorphous and metastable states (AMS) as well as those featuring a Grandjean texture (GJ) and a flow-aligned nematic state (FAN). The last two occur regardless of the initial state. They are for Blue Phase II BPII-1 (PRC) ($\dot{\gamma} \lesssim 3.91 \times 10^{-4}$, corresponding to $Er \lesssim 3.33$), BPII-2 (GJ) ($4.69 \lesssim \dot{\gamma} \lesssim 1.25 \times 10^{-3}$; $4 \lesssim Er \lesssim 10.67$), BPII-3 (FAN) ($1.875 \times 10^{-3} \lesssim \dot{\gamma}; 16 \lesssim Er$) and for Blue Phase I BPI-1 (AN) ($\dot{\gamma} \lesssim 1.95 \times 10^{-5}$; $Er \lesssim 0.17$), BPI-2 (PRC) ($3.91 \times 10^{-5} \lesssim \dot{\gamma} \lesssim 2.344 \times 10^{-4}$; $0.33 \lesssim Er \lesssim 2$), BPI-3 (AMS) ($3.125 \times 10^{-4} \lesssim \dot{\gamma} \lesssim 7.813 \times 10^{-4}$; $2.67 \lesssim Er \lesssim 6.01$), BPI-4 (GJ) ($9.375 \times 10^{-4} \lesssim \dot{\gamma} \lesssim 1.875 \times 10^{-3}$; $8 \lesssim Er \lesssim 16$) and BPI-5 (FAN) ($2.5 \times 10^{-3} \lesssim \dot{\gamma}, 21.34 \lesssim Er$).

As is standard^{16,35} disclination lines are represented by plotting an isosurface of the scalar order parameter q_s . Typical

choices are $q_s = 0.18$ for BPI and $q_s = 0.15$ for BPII. (Note that q_s is small but non-zero at the disclination core. The director is undefined there because the largest and second largest eigenvalues of \mathbf{Q} coincide.)

3.1 Blue Phase II

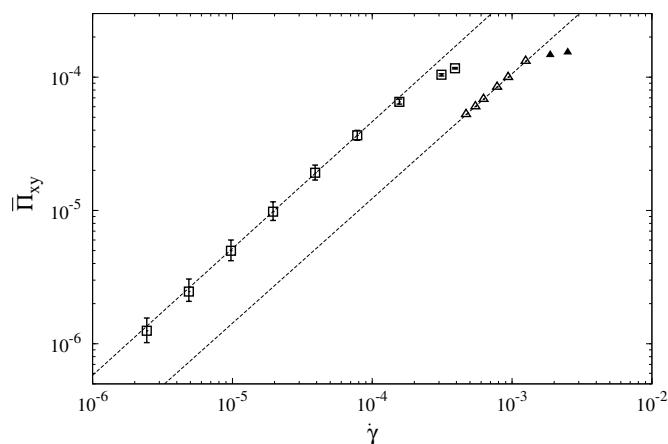


Fig. 1 Flowcurve $\bar{\Pi}_{xy}(\dot{\gamma})$ of BPII. The open squares show the average shear stress in the regime with periodically recurring conformations, whereas the error bars indicate the maximum and minimum stresses that occur during one cycle. Above a critical flow rate the network breaks up into a Grandjean texture (open triangles) or a flow-aligned nematic state at even higher flow rates (solid triangles). The two dashed lines represent fits to the data with $\bar{\Pi}_{xy}(\dot{\gamma}) \propto \dot{\gamma}^{0.95}$ (left) and $\bar{\Pi}_{xy}(\dot{\gamma}) \propto \dot{\gamma}^{0.94}$ (right), respectively.

We start our discussion with BPII as its flow behaviour is somewhat simpler than that of BPI. BPII has simple cubic symmetry and the disclination lines intersect and form a characteristic network of nodes. Ahead of the more detailed discussion we provide first a general overview of the flow behaviour at all applied shear rates.

Fig. 1 shows a flow curve, defined as time averages of the shear stress $\bar{\Pi}_{xy}$ as a function of shear rate $\dot{\gamma}$. For intermediate down to the lowest shear rates, the regime we refer to as BPII-1, a power law fit $\bar{\Pi}_{xy} = a\dot{\gamma}^b$ with $a = 0.30, b = 0.95$ describes the data to a very good approximation. This holds true even in regime BPII-2, where the network breaks up and the liquid crystal adopts a Grandjean structure. The coefficients of the power law fit are here $a = 0.68$ and $b = 0.94$. Hence, the degree of shear-thinning is remarkably small in BPII, in the range of shear rates which we have explored here.

Fig. 2 shows the ratio between the apparent viscosity, defined as $\eta_{app} = \langle \Pi_{xy} \rangle / \dot{\gamma} + \eta$, and the background viscosity η over total strain. A numerical value of $\eta_{app}/\eta = 1$ corresponds to a fully Newtonian flow, without any additional con-

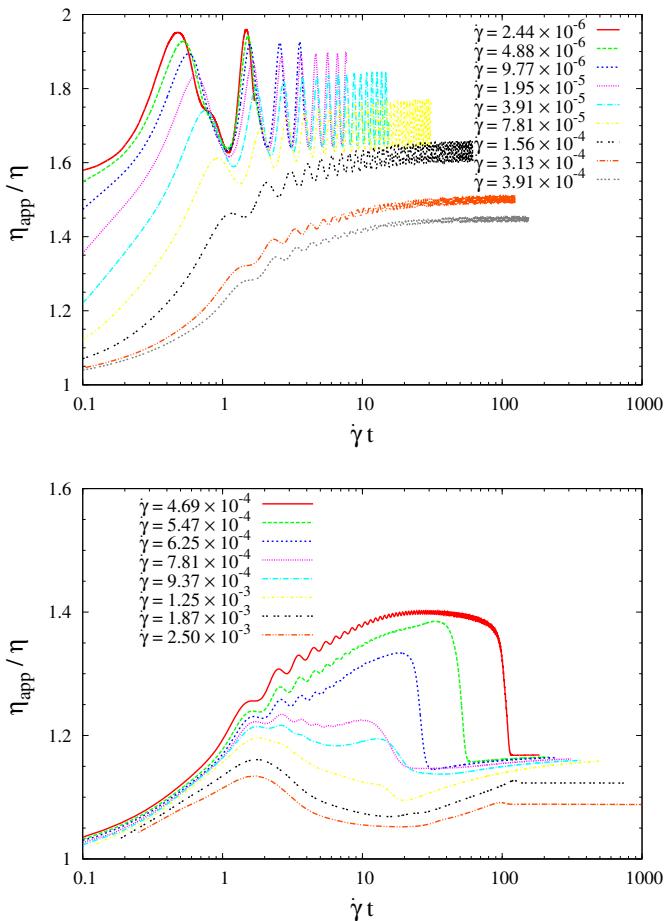


Fig. 2 Apparent viscosity $\eta_{app} = \Pi_{xy}/\dot{\gamma} + \eta$ (normalised to the background viscosity η) of BPII versus strain $\gamma = \dot{\gamma} \cdot t$ in the regimes BPII-1 (top, PRC) and BPII-2 (bottom, GJ) and BPII-3 (bottom, FAN). The value $\eta_{app}/\eta = 1$ corresponds to Newtonian flow without additional contribution of the liquid crystal.

tribution from the liquid crystal. The top picture shows data for regime BPII-1, where the periodic breakup and reconnecting of the network in shear flow causes sinusoidal oscillations in η_{app} . As noted in the Appendix, the absence at very low shear rate of a permeation mode is probably due to our choice of boundary conditions.

We observe another configuration, lying between those with periodically recurring conformations and the flow-aligned nematic state at shear rates $4.67 \times 10^{-4} \lesssim \dot{\gamma} \lesssim 1.25 \times 10^{-3}$ ($4 \lesssim Er \lesssim 10.67$). In this interval the network breaks up completely into a simple cholesteric liquid crystal, and oscillations in the stress signal are absent. The helical axis lies along the flow gradient direction (the so-called Grandjean texture¹) and the liquid crystal is flowing in “nematic planes” with the director field predominantly oriented in flow-vorticity plane.

The picture at the bottom of Fig. 2 gives data for this regime, to which we refer as BPII-2 and for BPII-3, the flow-aligned nematic state. We will address these regimes in section 3.3, as they are common to BPI and BPII.

The travelling helical waves which was predicted in^{21,22} when shearing a cholesteric helix along the vorticity axis turns out to be actually a metastable state and features larger stress and higher dissipation due to the tumbling motion of the local director field, which is absent in the Grandjean texture. For even higher shear rates the system undergoes a transition to a flow-aligned nematic state.

In the next sections we investigate the BPII-1 flow regime in more detail by looking at the kinetics of the disclination network.

3.1.1 Regime BPII-1: low and intermediate shear rates

Fig. 3 shows the disclination network in shear flow as it undergoes homogeneous shearing in the BPII-1 flow regime. The disclination lines break up and reconnect further downstream, forming a periodically recurring pattern. The period needed for a pattern to break up and reform along the flow direction is $\tau_F = 1/\dot{\gamma}$.

The general appearance of the flowing network is, apart from the homogeneous distortion, very close to that of the quiescent blue phase at equilibrium. This holds for all shear rates in the BPII-1 regime.

Interestingly, while being displaced with the flow the entire network moves along the vorticity direction (z) as well. A similar behaviour has been recently observed for blue phases in shear flow in confined geometries³⁵. In contrast to the stick-slip motion that has been reported there, in this case the movement is steady. The vorticity motion occurs in such a way that the positions of breakup and reconnection point in the network, visible in Fig. 3, are slightly offset and allow the network to travel along the z -direction. The periodicity of the motion along the vorticity direction is $\tau_V = 6\tau_F$, i.e. it takes a displacement of six unit cells along x (the flow direction)

for the network to move one unit cell along z (the vorticity direction).

Fig. 4 shows the disclination network in the middle of a breakup-reconnection cycle, with superimposed velocity vectors. The plot shows the secondary velocity components, obtained by projecting the velocity onto a plane perpendicular to the flow direction. This gets rid of the dominating velocity component along the flow, v_x , and allows to visualise the patterns in the two much smaller components v_y and v_z . The magnitude of the secondary components is typically in the range of a few percent of the primary flow component, for the shear rates and system sizes simulated here.

Characteristic bands are visible in Fig. ??, which are oriented along the vorticity direction. The direction of the flow in each of the bands depends on the helicity of the underlying cholesteric liquid crystal (i.e. on the sign of q_0 in Eq. 1), and the velocity patterns for left-handed and right-handed BPs are one the mirror image of the other. Further quantitative evidence for a direct link between the sense of motion and the helicity can be gained by time-averaging over individual cycles.

Table 1 in the Appendix gives minima, maxima, averages and standard deviations of the velocity components. All values for the two runs with inverted helicity are identical apart from a change of sign in the z -components. There is only a slight imbalance in magnitude between the maximum and minimum velocities along the z -direction. Therefore the averaged flow along the vorticity direction, leading to network motion, is very small, and much smaller than the local velocity flow. This suggests that the flow of the network along the vorticity direction is permeative (i.e. the fluid moves but the network as a whole moves much less).

3.2 Blue Phase I

BPI has body-centred cubic symmetry and, unlike BPII, the disclination lines characterising its equilibrium structure are well separated and do not intersect. (The quiescent state resembles the first frame in Fig. 9 below.) We believe that this topological difference is responsible for most of the differences between BPI and BPII regarding their rheological response. We present again key aspects in an overview before we address specific features in more detail.

Fig. 6 shows the ratio between the apparent viscosity η_{app} and η versus time and strain for the same shear rates as in Fig. ???. The plot shows that there is more shear thinning in BPI than in BPII, for the shear rate range we considered. Through much of the flow range we studied, the average shear stress versus shear rate can be fitted reasonably well by a power law $\bar{\Pi}_{xy} = a\dot{\gamma}^b$ with $a = 0.019, b = 0.63$.

For intermediate (but not low) shear rates, we once more find regular oscillations in the stress response versus time, and

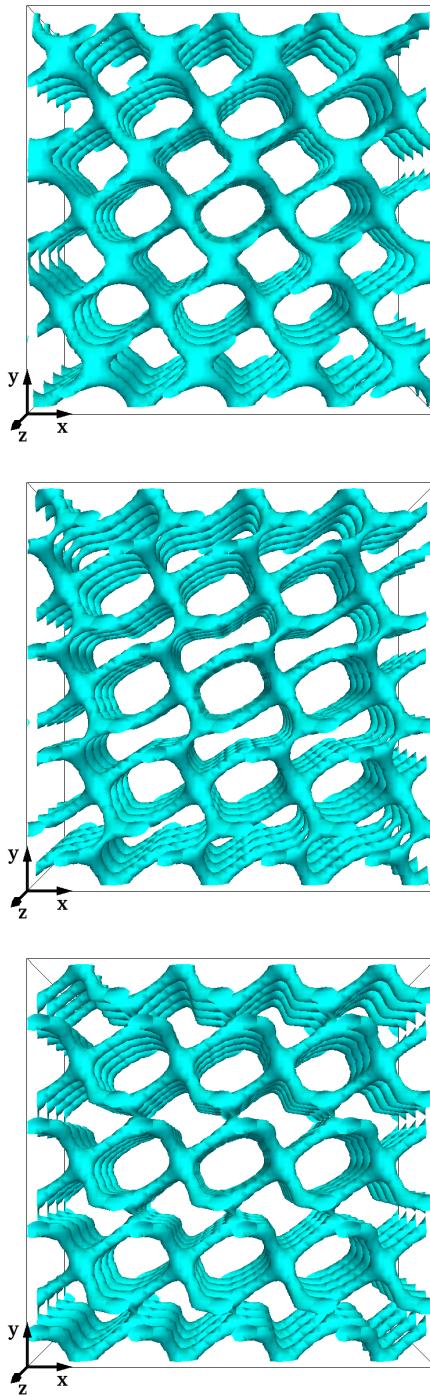


Fig. 3 Disclination network of BPII in shear flow: The pictures show a typical sequence of snapshots in the steady state at $\dot{\gamma} = 1.56 \times 10^{-4}$ and time steps $t = 1.60, 1.64, 1.65 \times 10^5$. The velocity gradient is oriented along the vertical direction (y), whereas the horizontal direction (x) is the flow direction. Lees-Edwards boundary conditions have been imposed in such a way that the network moves to the right in the upper half and to the left in the lower half of the picture.

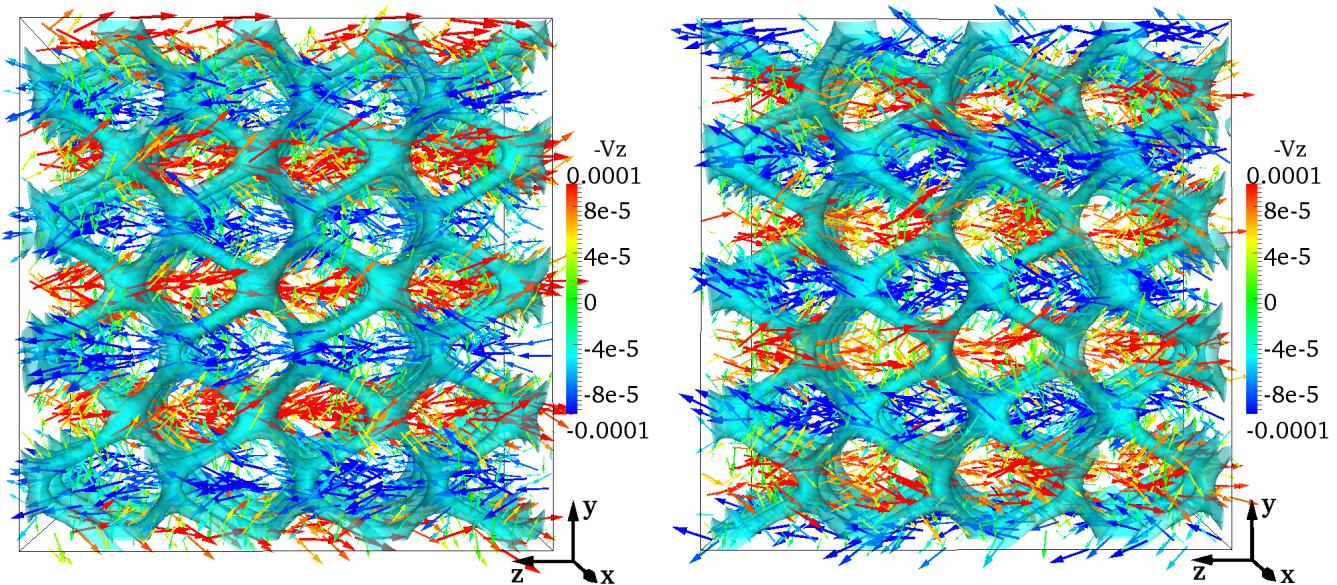


Fig. 4 Velocity patterns and disclination network in BPII for positive (left) and negative (right) helicity of the underlying cholesteric helix: The pictures show velocity vectors $(0, v_y, v_z)$. The view is along the x -direction. This means the velocity \mathbf{v} has been projected onto a plane perpendicular to the flow direction. The vertical and horizontal direction are the gradient and vorticity direction, respectively. The colour code gives the magnitude and sign of the component in vorticity direction. The snapshot shows a typical frame during a periodically recurring sequence. The network on the left with right-handed helicity ($q_0 > 0$ in Eq. 1) travels rightwards, whereas the one on the right, with reversed helicity, moves leftward. The periodicity of the motion along the vorticity direction is six times longer than the time it takes the network to reconnect along the flow direction.

in the network morphology – we label this regime BPI-2. The BPI-2 regime now does not persist to arbitrarily low shear: there the oscillations become irregular and seemingly chaotic, and after a few cycles the disclination pattern becomes amorphous. We refer to this flow regime as BPI-1. The BPI-2 regime is also unstable if the shear is increased *above* a critical value – close to this breakup point the oscillations in the shear stress show an increasingly complex pattern, until the network breaks up completely and a new regime, BPI-3, is entered. In regime BPI-3 ($6.25 \times 10^{-4} \lesssim \dot{\gamma} \lesssim 7.81 \times 10^{-4}$; $10.67 \lesssim Er \lesssim 13.33$) we observe that the BPI network transforms under shear into a quasi two-dimensional state consisting of double-twist rolls. Finally, for shear rates such that $9.37 \times 10^{-4} \lesssim \dot{\gamma} \lesssim 1.25 \times 10^{-3}$ ($16 \lesssim Er \lesssim 21.33$) we observe the same cholesteric configuration with the helical axis along the gradient direction as reported above for BPII-2 (see section 3.3).

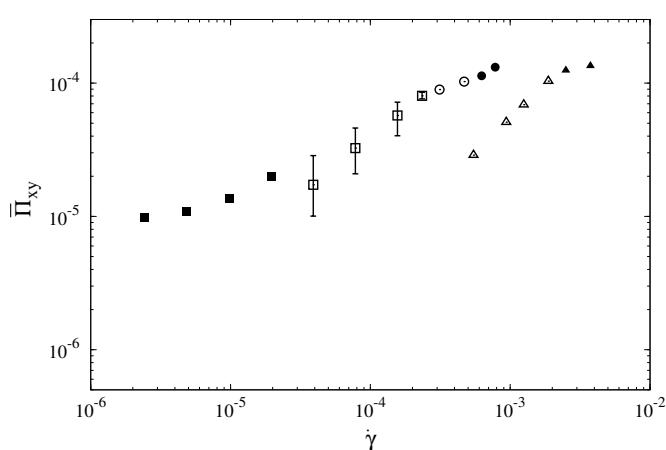


Fig. 5 Flow curve $\bar{\Pi}_{xy}(\dot{\gamma})$: Depending on the configuration in steady shear flow five different regimes can be identified: amorphous BP network with yield stress (BPI-1, solid squares), steady flow with periodically recurring patterns (BPI-2, open squares), breakup of the disclination network into amorphous state (BPI-3, open circles) and metastable configurations (BPI-3, solid circles), cholesteric helix with axis in gradient direction (BPI-4, open triangles) and flow-aligned nematic state (BPI-5, solid triangles). The error bars represent the minimum and maximum shear stress during one cycle in regime BPI-2.

3.2.1 Regime BPI-1: low shear rates The rheological response of BPI at low shear rate, $\dot{\gamma} \lesssim 1.95 \times 10^{-5}$ ($Er \lesssim 0.33$), is strikingly different from that of BPII and appears to show a yield stress. An explanation for this behaviour can be found by looking more closely at the average shear stress (where the background viscosity contribution has been subtracted) and free energy density as a function of time, as shown in Fig.

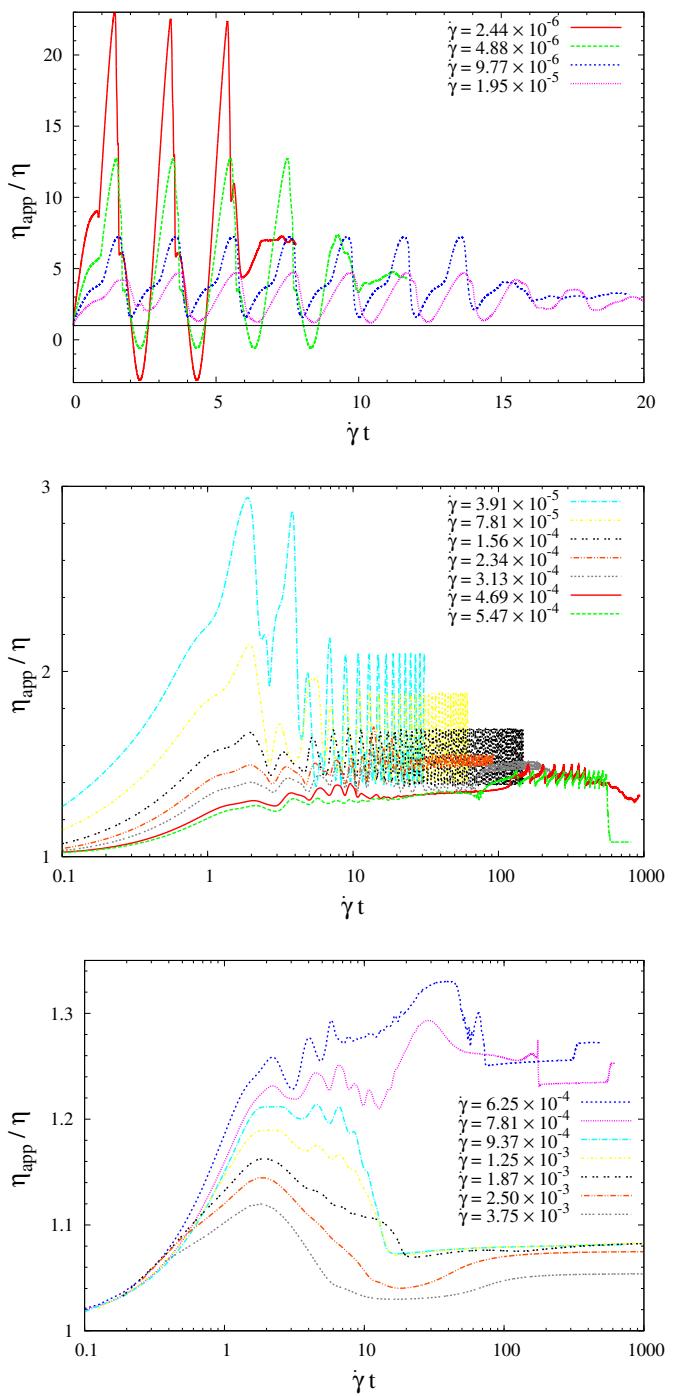


Fig. 6 Apparent viscosity $\eta_{app} = \langle \bar{\Pi}_{xy} \rangle / \dot{\gamma}$ (normalised by η) of BPI versus time (top) and strain (bottom).

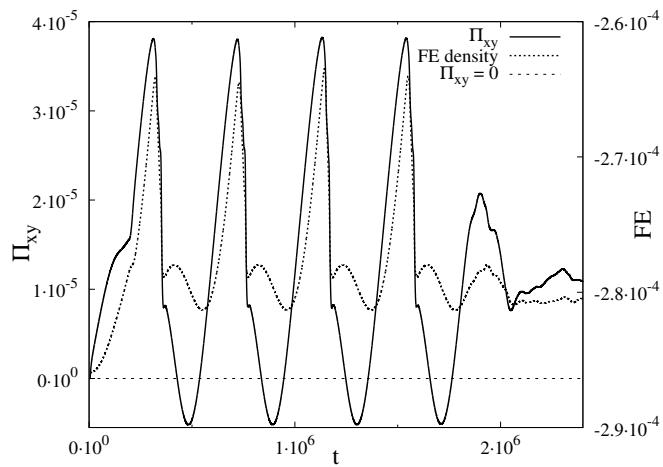


Fig. 7 Average thermodynamic shear stress and free energy density of BPI at shear rate $\dot{\gamma} = 4.88 \times 10^{-5}$, $Er = 0.08$: The negative branch in the stress is related to a local maximum and a following local minimum in average free energy density. This creates unstable conditions that lead to reconstruction of the defect lattice into an amorphous network state which seemingly features a yield stress.

7. When the quiescent and equilibrated BPI network begins to flow the shear stress increases steeply and goes through a maximum. Shortly after it goes negative (the total shear stress with the background viscosity contribution remains positive) to become positive again later on, forming a complete cycle. The amplitude of the excursions in these stress oscillations, and the presence of a part of the period where the thermodynamic contribution to the stress is negative, suggest that the BPI network is subject to large forces. Seemingly, these eventually cause the flow-induced collapse to an amorphous network state with an apparent yield stress. We should underscore that this low shear regime might depend on our choice of boundary conditions which in practice are equivalent to having infinitely distant walls to which the network is anchored (see discussion in the Appendix). With free boundary conditions and for very low wall velocity, permeative flows might instead allow for some slip of the BPI network, as in cholesterics sheared along their axis²⁰.

To gain further insight into the flow-induced deformation, and eventual breakdown, of BPI, it is instructive to follow the dynamics of the network under a slow flow – this is depicted in Fig. 9. Three different stages can be distinguished. Just after the onset of the shear flow, the disclination lines in BPI get more and more squeezed together, and this is incompatible with the defect topology in this phase. Consequently, the network adopts a flow-induced conformation which consists of intertwined helices that stretch during the shear transformation. This helical conformation that emerges already at strains $\gamma \approx 1$ is shown in Fig. 9 (top row). At this point the original

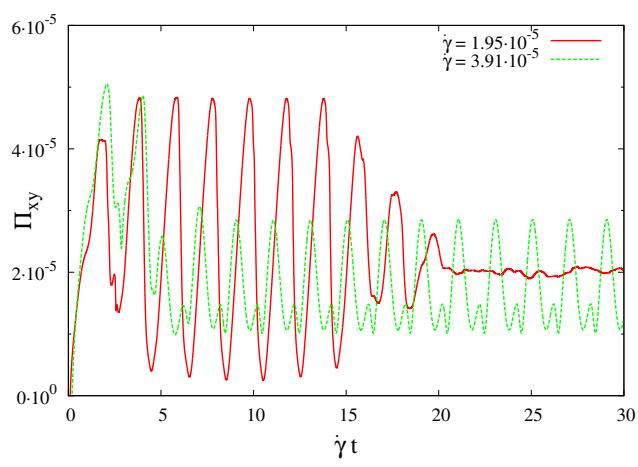


Fig. 8 Thermodynamic shear stress Π_{xy} versus strain near the transition between regime BPI-1 and BPI-2. The shear rates are $\dot{\gamma} = 1.95 \times 10^{-5}$ (red solid) and $\dot{\gamma} = 3.91 \times 10^{-5}$ (green dashed). Compared to lower shear rates in BPI-1 where the stress goes negative (Fig. 7) the stress is throughout positive, but exhibits very large fluctuations during one cycle, which are absent in regime BPI-2 at higher shear rates than shown here.

BPI has already been significantly deformed. Shortly afterwards regular oscillations set up temporarily, where the disclinations form double helices which tilt and realign under the shear. There is no perceivable movement of the network along the vorticity direction at any stage of the cycle. (This contrasts with the BPII-2 results reported earlier.)

This mode of flow proves unstable, as after a few cycles distortions appear which lead to further destabilisation – presumably in view of the large stress fluctuations discussed above. Finally, the system loses order and transforms into an amorphous network state with almost constant stress in the region of $\Pi_{xy} \approx 1 \times 10^{-5}$ LBU. If the shear is stopped, this flow-induced amorphous state remains arrested and metastable, and cannot find its way back to the original BPI structure.

3.2.2 Regime BPI-2: intermediate shear rates Adjacent to regime BPI-1 ($\dot{\gamma} \lesssim 1.95 \times 10^{-5}$; $Er \lesssim 0.33$) but at slightly larger shear rates ($3.91 \times 10^{-5} \lesssim \dot{\gamma} \lesssim 4.687 \times 10^{-4}$; $0.67 \lesssim Er \lesssim 8$) lies another region where the network flows with periodically recurring conformations (Fig. 6). We refer to this region as BPI-2. The transition between BPI-1 and BPI-2 is clear when looking at Fig. 8 which shows the shear stress versus total strain. A qualitative difference between these two is the absence in BPI-2 of the large stress fluctuations that occur during the early cycles in regime BPI-1. (Recall that for even lower $\dot{\gamma}$ the thermodynamic shear stress becomes temporarily negative (Fig. 7), which caused destabilisation of the periodic network and led to the amorphous configuration in the steady state.) Hence, an explanation for the existence

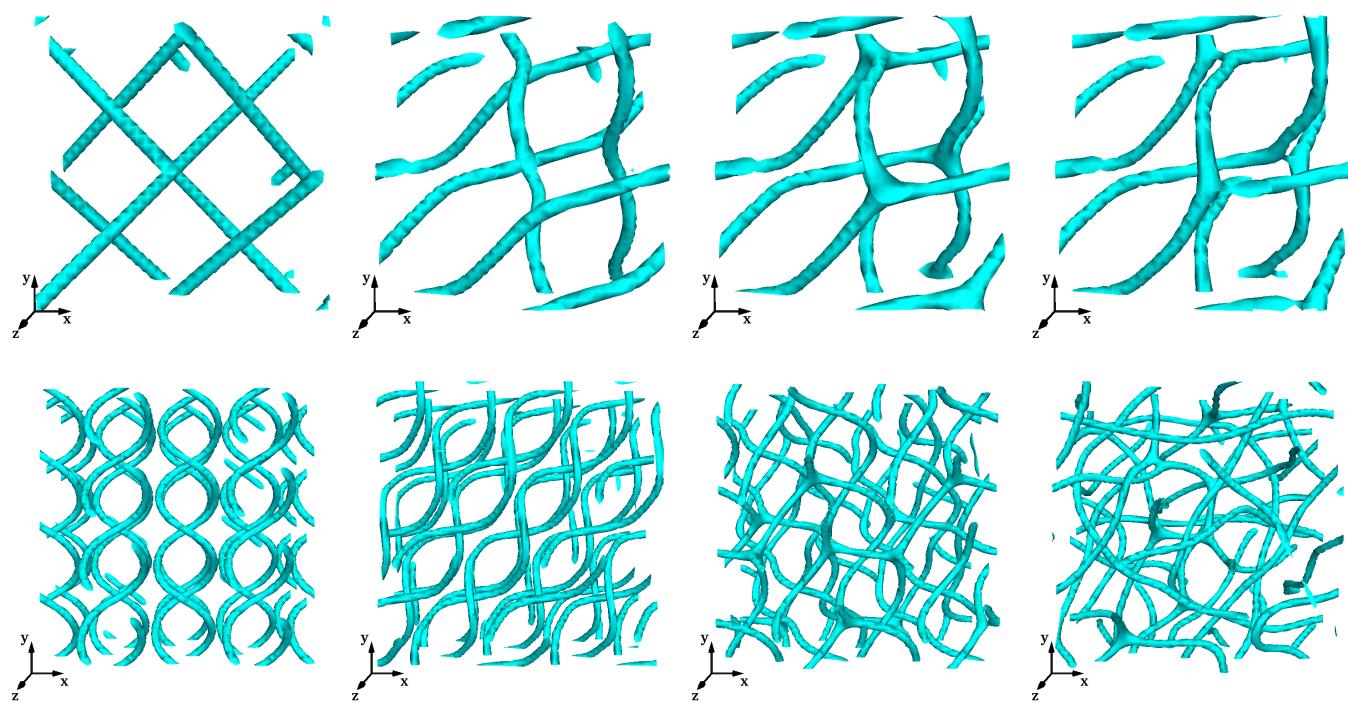


Fig. 9 Snapshots of BPI disclination network at $\dot{\gamma} = 4.88 \times 10^{-6}$: Depicted is the transition from the quiescent state to flow-induced, intertwined helices that undergo a recurring structural transformation. The top row shows a section of one unit cell for early times $t = 1 \times 10^4, 1.8 \times 10^5, 2.0 \times 10^5$ and 2.1×10^5 . The bottom pictures on the far left, centre left and centre right show the situation at later time steps $t = 1.65, 1.8$ and 2.0×10^6 , which the system passes for several cycles before it ends up in an amorphous state (bottom row far right, at time step $t = 2.4 \times 10^6$).

of the regular BPI-2 oscillations could be that these are fast enough to bypass or suppress large stress fluctuations – this leads to a different order structure and to a stabilisation compared to the oscillations found transiently in the BPI-1 regime.

Fig. 10 shows snapshots of the periodically recurring BPI in steady shear flow. Contrary to BPII-1 at these shear rates, BPI-2 does not resemble the equilibrium configuration undergoing a homogeneous topology-conserving transformation. It features rather intricate, flow-induced conformations. Surprisingly, if the shear is switched off in the BPI-2 regime at some point during an oscillation, the flow-induced configuration usually reverts to a perfect quiescent BPI – in stark contrast with what happens in BPI-1, for lower shear rates.

Finding the recurrence period for BPI is not simple, as this depends subtly on the applied shear rate. The analysis is complex for a number of reasons. First, the recurrence period for the stress is not always the same as that of the network configuration. For instance, at $\dot{\gamma} = 3.9062500 \times 10^{-5}$, it takes $2/\dot{\gamma}$ for one stress cycle to complete, but $4/\dot{\gamma}$ for the network to recur – this is because the configurations at times t and $t + 2/\dot{\gamma}$ are mirror-images. This is by no means generic though: for example at $\dot{\gamma} = 4.8828125 \times 10^{-5}$ it takes $8/\dot{\gamma}$ for one stress cycle to complete, but $16/\dot{\gamma}$ for the network to recur. For some higher shear rates, the recurrence period of stress and network conformations are instead the same. Second, there exist isolated values (or perhaps small ranges) of shear rates for which the recurrence period is much longer and far more irregular, with seemingly intermittent dynamics (e.g. at $\dot{\gamma} = 1.3671875 \times 10^{-4}$ the network configuration repeats itself after $6/\dot{\gamma}, 12/\dot{\gamma}, 22/\dot{\gamma}$). Taken together, all this evidence very much suggests that the BPI-2 range includes at least some instances of “rheochaos”^{36,37}.

Fig. 11 depicts a snapshot of the secondary velocity components v_y and v_z for left- and right-handed chirality of the blue phase. The emerging pattern is similar to that of BPII from Fig. ??, but differs in that there is no simple separation into bands with opposite secondary velocity. However, just as in the case of BPII, both patterns are mirror images and feature the highest velocities in the regions where the largest conformational changes take place.

3.2.3 Regime BPI-3: high shear rates

3.3 Transition to cholesteric helix and flow-aligned nematic state

At shear rates beyond the regimes BPI-3 and BPII-2, but below the transition to a flow-aligned nematic state at still higher shear rates, we found another regime where both blue phases adopt the same configuration in steady shear flow, independent of their initial state. The director field of this configuration is shown in Fig. 13. It consists of a simple cholesteric helix with the helical axis oriented along the velocity gradient direction.

The state is translational invariant along the flow and vorticity direction and therefore also one-dimensional. However, in contrast to the travelling helical wave of BPII-2 the director field is now static and there is no tumbling motion. While flowing the director retains its relative orientation. Consequently, the shear stress is lower than for the 2D double-twist rolls of BPI-3 and the travelling helical wave (Fig. ?? and 6) as there is less dissipation.

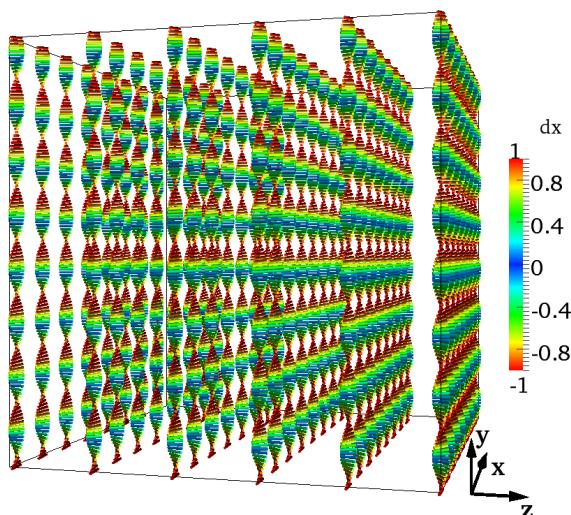


Fig. 13 Flow induced cholesteric state. The orientation of the helix is along the gradient direction. The director field is quasi-static during the flow, which occurs in “nematic layers”. Colour coding indicates the x component of the director field (see colorbar on the right). For clarity only selected sites are shown along the y -direction (along which the structure is translationally invariant).

4 Conclusions

In summary, our work constitutes the first large scale simulation of bulk flow behaviour of cubic BPs in simple shear flow.

We were able to characterise the rheology of cubic BPs, and identified two different flow regimes for BPII and three different ones for BPI. Below an Erickson number of $Er \simeq 3$, BPII exhibits weak shear-thinning and obeys a power-law flow curve with exponent close to unity. The BPII disclination network breaks up and reconnects in the flow, which leads to a periodically recurring dependence of the shear stress. The flow-induced conformation looks generally very similar to the quiescent network at equilibrium. While being homogeneously transformed due to the shear flow the disclination network moves steadily in the vorticity direction apparently by a permeation mechanism. The sense of motion is directly linked to the helicity of the underlying cholesteric phase.

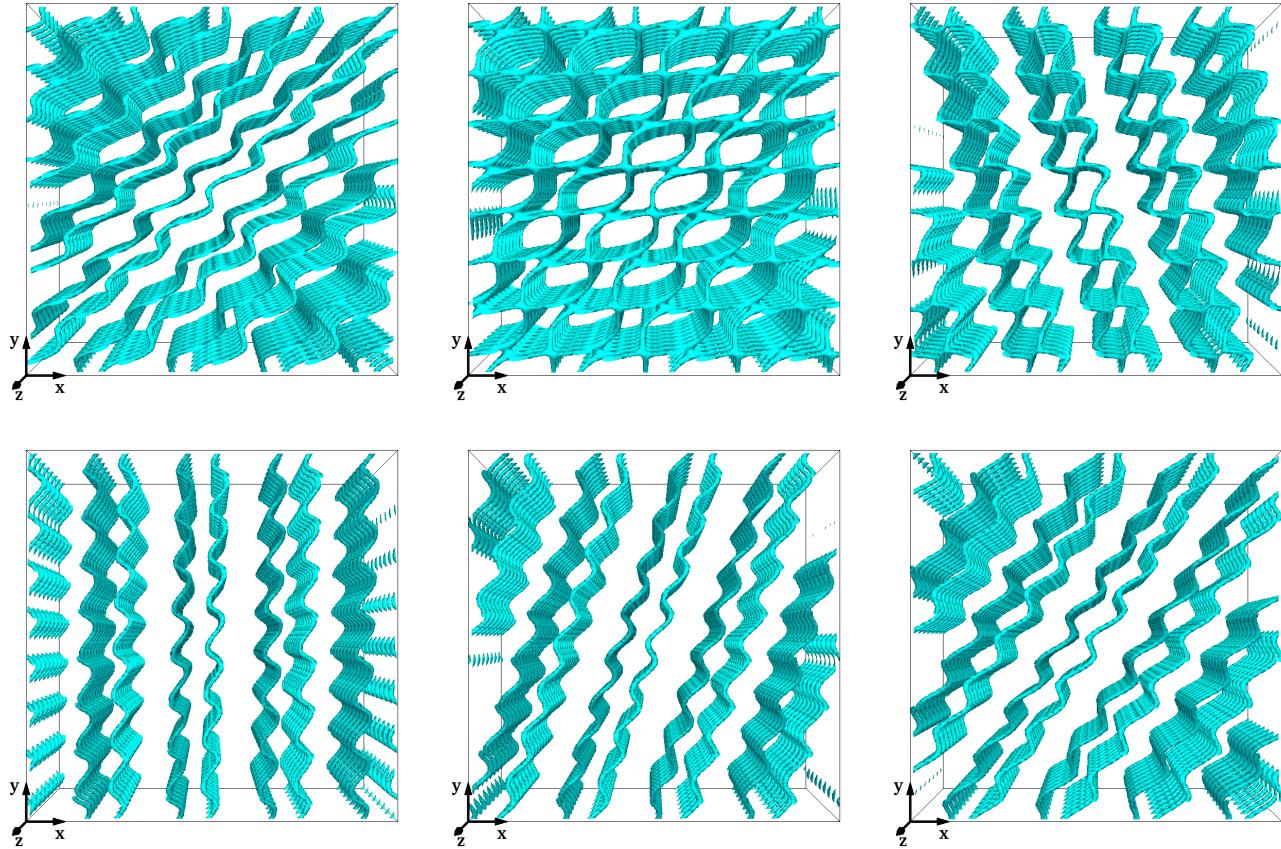


Fig. 10 Disclination network of BPI in regime BPI-2 (intermediate flow rates): The sequence shows a typical cycle of shear-induced transformations in the steady state at $\dot{\gamma} = 1.56 \times 10^{-4}$ and time steps $t = 3.64, 3.66, 3.68, 3.70, 3.72, 3.74 \times 10^5$ in flow-gradient plane. During every cycle the network is displaced also along the gradient and vorticity direction.

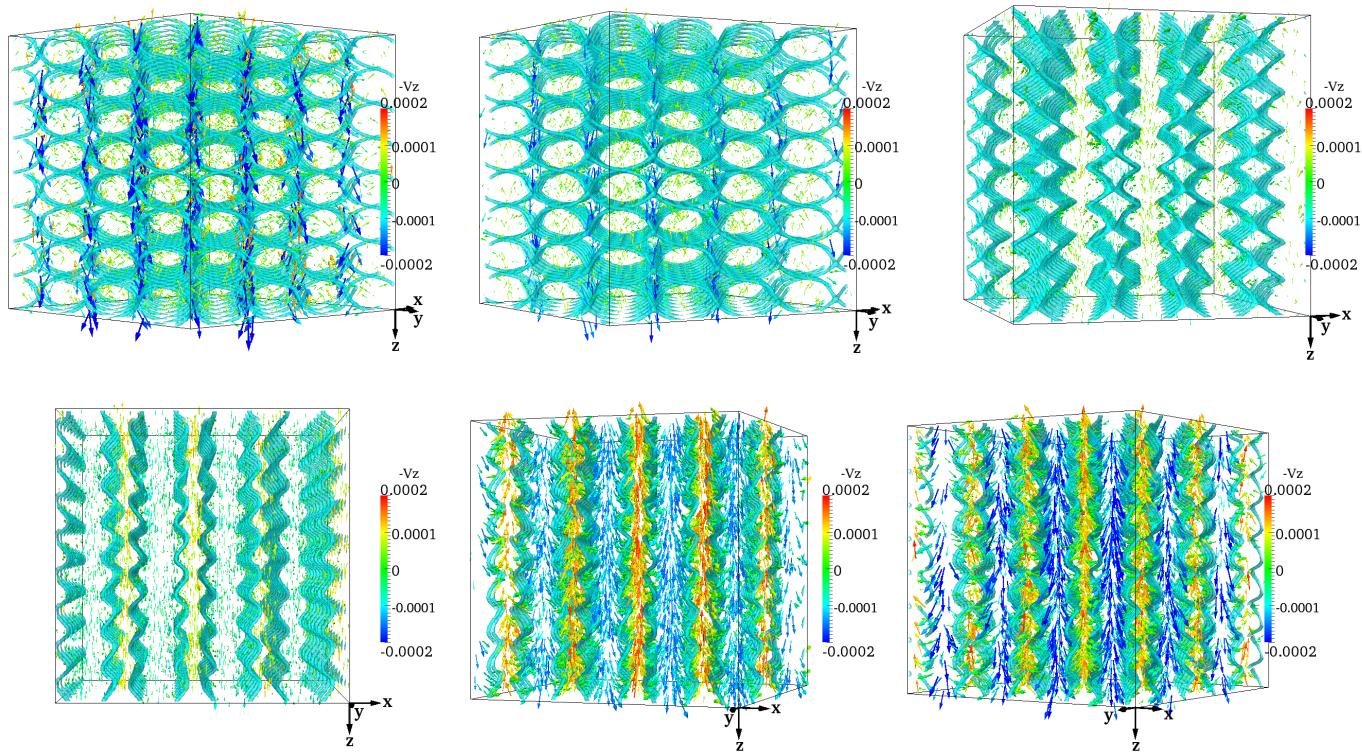


Fig. 11 Velocity patterns in BPI for positive helicity: The pictures show a snapshot of the periodically recurring patterns in the secondary velocity components (v_y, v_z) at $\dot{\gamma} = 1.56 \times 10^{-4}$ and time steps $t = 3.64, 3.66, 3.68, 3.70, 3.72, 3.74 \times 10^5$, i.e. for the same shear rate and time steps as in Fig. 10. The colour code gives the magnitude and sign of the z -component. For negative helicity the sense of motion of the network and the secondary velocity components are inverted just as in Fig. ???. Note that the viewing direction has been adapted in each snapshot to give the clearest view on the band-like pattern between the inclination lines.

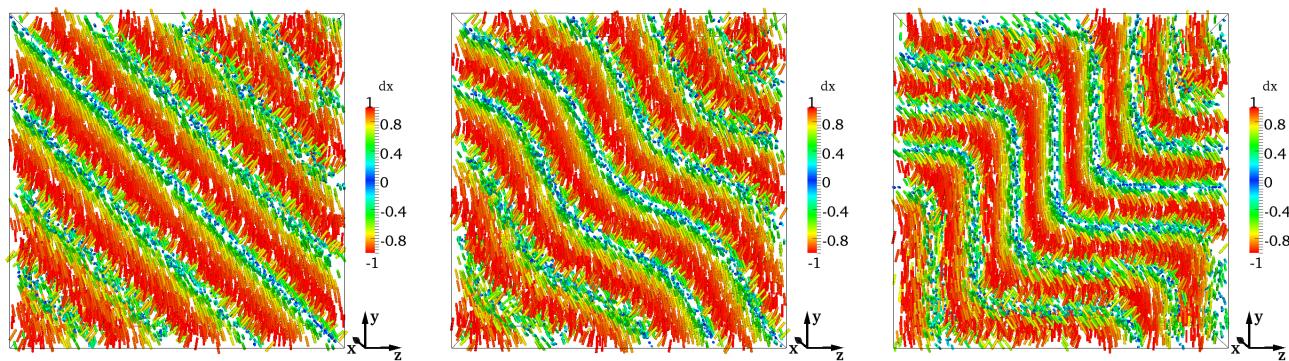


Fig. 12 Director field of BPI at high shear rate: The pictures show a region of $L_x \times L_y \times L_z = 32^3$ lattice sites with the flow/gradient direction oriented along the horizontal/vertical axis. The colour code gives the magnitude of the z -component of the director field. The data show that the structure is quasi-2D, with rolling double twist patterns in the xy plane, and an approximate translational symmetry along the vorticity direction.

At larger Ericksen numbers, $4 \lesssim Er \lesssim 10$, the flowing BPII network breaks up in a two-step manner. First a simple cholesteric helix forms with the helical axis along the vorticity direction. The flow excites a travelling helical wave, as predicted theoretically in Ref.^{21,22}. Upon increasing the shear, the helix rotates and orients along the gradient direction, where it can support larger flow rates due to the translational symmetry along the flow direction. Still larger flow rates break this residual cholesteric order and leave a standard flow-aligned nematic state, at the highest shear rate studied.

Interestingly, BPI shows a flow behaviour that is very different to that of BPII. This is a direct consequence of the topological differences between their disclination networks. Below an Ericksen number of $Er \sim 0.4$, the BPI network cannot both flow and retain a regular appearance. It eventually reorganises into an amorphous network that features yield-stress behaviour. The apparent reason is that shortly after the onset of the shearing very large stress fluctuations occur. At the lowest shear rates the thermodynamic contribution to the shear stress becomes temporarily negative during each cycle. These fluctuations seem to destabilise the network and eventually trigger the transition into an amorphous state with a residual yield stress.

At slightly larger Ericksen numbers $0.4 \lesssim Er \lesssim 8$, BPI kinematically bypasses the large stress fluctuations and flows with very complex, periodically recurring flow-induced conformations. These conformations entail regularly arranged helical disclinations which rotate due to the shear – a new flow-induced state which was not previously predicted. The oscillatory dynamics is in some cases intermittent, and the recurrence period for the network conformations and stress patterns is not given by a simple formula as for BPII. Despite their complex appearance, the flow-induced conformations are topologically connected to the quiescent BPI and after switching off the shear flow a defect-free blue phase reforms. It is tempting to interpret the unsteady oscillations seen in this regime as an instance of deterministic rheological chaos^{37,38}.

Just as in the case of BPII, the BPI network breaks up at larger Ericksen numbers in a two-step process. First, at $10 \lesssim Er \lesssim 15$, it adopts a quasi-two-dimensional configuration that is translationally invariant along vorticity direction. Then, at $16 \lesssim Er \lesssim 22$, it forms a cholesteric helix along the flow gradient direction. Finally, at $Er \gtrsim 30$ (BPI) and $Er \gtrsim 10$ (BPII) the configuration is a flow-aligned nematic state. Although experimental evidence to support our results is currently not available, we hope this work will inspire such experiments, and believe it can shed some light on the flow properties of complex liquid-crystalline phases.

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Appendix

In this Appendix we give some more details on our simulations, including the relationship between boundary conditions and permeation flow.

Table 1 lists the parameters chosen for our runs (values of the reduced temperature and reduced chirality are given in the caption), together with the minima, maxima, and standard deviations of the velocity field in the three directions (discussed further in the main text). The first column also describes what flow regime each simulation leads to in steady state.

We close with a technical note on the choice of boundary conditions. In all our runs we used Lees-Edwards boundary conditions: this is the sheared equivalence of periodic boundary conditions, for both the velocity and the order parameter field. While extremely useful to simulate bulk flow, we note that our boundary conditions impose macroscopic distortion of the network, and are effective analogous to infinitely distant but anchored wall along the xy plane. In other words, due to the periodic boundary conditions along the flow gradient direction, there can be no slip in the network deformation (or strain) at any point. Effectively, this disallows permeation flows, which on the other hand require a non-zero velocity field and, simultaneously, much lower (ideally zero) perturbations of the blue phase network. Consequently our results are effectively valid for sheared bulk BPs which are anchored at the walls. In practice, permeation flows are only restricted to very low flow rates, and are unstable for intermediate and fast flows²⁵, where the response of the network depends less on the details of the boundary conditions used at the wall.

	$\dot{\gamma}$	Er	q_0	$\bar{v}_{x,min}$	$\bar{v}_{x,max}$	$\bar{v}_{x,std}$	$\bar{v}_{y,min}$	$\bar{v}_{y,max}$	$\bar{v}_{y,std}$	$\bar{v}_{z,min}$	$\bar{v}_{z,max}$	$\bar{v}_{z,std}$
BPI				$\times 10^5$								
BPI-1 (AN)	0.24	0.02	0.1388									
BPI-1 (AN)	0.49	0.04	0.1388									
BPI-1 (AN)	0.98	0.08	0.1388									
BPI-1 (AN)	1.95	0.17	0.1388									
BPI-2 (PRC)	3.91	0.33	0.1388									
BPI-2 (PRC)	7.81	0.67	0.1388									
BPI-2 (PRC)	15.63	1.33	0.1388									
BPI-2 (PRC)	15.63	1.33	-0.1388									
BPI-2 (PRC)	23.44	2.00	0.1388									
BPI-3 (AMS)	31.25	2.66	0.1388									
BPI-3 (AMS)	46.87	4.00	0.1388									
BPI-3 (AMS)	54.69	4.67	0.1388									
BPI-3 (AMS)	62.50	5.33	0.1388									
BPI-3 (AMS)	78.13	6.01	0.1388									
BPI-4 (GJ)	93.75	8.00	0.1388									
BPI-4 (GJ)	125.0	10.66	0.1388									
BPI-4 (GJ)	187.5	16.00	0.1388									
BPI-5 (FAN)	250.0	21.33	0.1388									
BPI-5 (FAN)	375.0	32.00	0.1388									
BPII				$\times 10^5$								
BPII-1 (PRC)	0.24	0.02	0.1963	-7.8	7.7	0.1	-0.2	0.2	0.1	-0.3	0.3	0.2
BPII-1 (PRC)	0.49	0.04	0.1963	-15.5	15.4	0.2	-0.3	0.4	0.2	-0.5	0.4	0.2
BPII-1 (PRC)	0.98	0.08	0.1963	-31.0	30.8	0.3	-0.6	0.6	0.4	-0.9	0.8	0.4
BPII-1 (PRC)	1.95	0.17	0.1963	-62.0	61.6	0.6	-1.2	1.2	0.8	-1.8	1.6	0.8
BPII-1 (PRC)	3.91	0.33	0.1963	-123.9	123.3	1.2	-2.1	2.1	1.6	-3.4	3.0	1.5
BPII-1 (PRC)	7.81	0.67	0.1963	-247.5	246.8	2.0	-3.8	3.8	3.0	-6.5	5.5	2.6
BPII-1 (PRC)	15.63	1.33	0.1963	-494.3	493.7	2.5	-5.5	-5.5	4.0	-10.9	8.7	3.9
BPII-1 (PRC)	15.63	1.33	-0.1963	-494.3	493.7	2.5	-5.5	5.5	4.0	-8.7	10.9	3.9
BPII-1 (PRC)	31.25	2.67	0.1963	-987.5	987.1	3.5	-6.9	6.9	5.1	-17.7	13.1	5.0
BPII-1 (PRC)	39.06	3.33	0.1963	1233.9	1233.6	3.7	-6.9	6.9	5.1	-20.0	14.5	5.2
BPII-2 (GJ)	46.87	4.00	0.1963	-1479.8	1477.0	-	-	-	-	-10.0	9.6	-
BPII-2 (GJ)	54.69	4.67	0.1963	-1720.5	1720.8	-	-	-	-	-11.5	11.2	-
BPII-2 (GJ)	62.5	5.33	0.1963	-1966.7	1966.2	-	-	-	-	-13.3	12.7	-
BPII-2 (GJ)	78.13	6.01	0.1963	-2458.3	2457.8	-	-	-	-	-16.9	15.7	-
BPII-2 (GJ)	93.75	8.00	0.1963	-2949.7	2949.7	-	-	-	-	-20.3	18.4	-
BPII-2 (GJ)	125.0	10.67	0.1963	-3932.8	3932.8	-	-	-	-	-26.6	22.4	-
BPII-3 (FAN)	187.5	16.00	0.1963	-5906.2	5906.2	-	-	-	-	-0.6	0.4	-
BPII-3 (FAN)	250.0	21.33	0.1963	-7874.9	7874.9	-	-	-	-	-0.5	0.3	-

Table 1 Minima, maxima and standard deviation of time-averaged velocity components for BPI ($\tau = -0.5, \kappa = 1.0$) and BPII ($\tau = -0.5, \kappa = 2.0$) in steady shear flow. All velocity values are given in 10^{-5} lattice units. The transient dynamics has not been included in the averages. The regimes comprise the formation of an amorphous network (BPI-1), periodically recurring conformations with oscillatory response (BPI-2 and BPI-1), amorphous and metastable steady states (BPI-3), a Grandjean texture (BPII-2 and BPII-4), and a flow-aligned nematic state (BPI-5 and BPII-3). (Last six columns refer to the secondary flow.)

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