Blue phases of cholesteric liquid crystals offer a spectacular example of naturally occurring disclination line networks. Here we numerically solve the hydrodynamic equations of motion to investigate the response of three types of blue phases to an imposed Poiseuille flow. We show that shear forces bend and twist and can unzip the disclination lines. Under gentle forcing the network opposes the flow and the apparent viscosity is significantly higher than that of an isotropic liquid. With increased forcing we find strong shear thinning corresponding to the disruption of the defect network. As the viscosity starts to drop, the imposed flow sets the network into motion. Disclinations break-up and re-form with their neighbours in the flow direction. This gives rise to oscillations in the time-dependent measurement of the average stress.

61.30.Mp,83.50.-v,83.80.Xz

In many cholesteric liquid crystals the transition between the regular cholesteric phase and the isotropic phase occurs through one or more intermediate phases known as “blue phases” (BPs) [1–3]. Understanding the structure of the blue phases was a theoretical and experimental tour de force clearly summarised in [3].

Blue phases occur because the locally favoured state is a double twist (i.e. a rotation of the director field in two directions, as opposed to the single twist of the cholesteric) which is globally incompatible with the requirement of continuity [3]. This results in the formation of networks of defects that separate local regions of double twist with axes aligned in different directions.

The networks can be intricate and lead to structures that are periodic at large length scales. For example the BPI and BPII phases are 3-dimensional cubic phases where the orientational order can be periodic on the scale of the wavelength of visible light. Due to this, they have potential applications in fast light modulators [4], tunable photonic crystals [5] and lasers [6]. The main factor limiting the technological exploitation of BPs is that they are generally only stable over a temperature range ∼ 1 K. However recently it has been shown that the addition of polymers can stabilise BPI over more than 60 K opening perspectives in BP technology.

Though the static structure of BPs is well accepted essentially nothing is known of their rheology. Given the intricacy of the director configurations one might expect that elastic and viscous responses to an external stress combine to give highly non-Newtonian behaviour under flow. Cholesterics show a strong dependence of viscosity on stress and, in particular, a striking permeation mode where the backflow acts to minimise director distortions [8]. In blue phases there is also a fully three-dimensional network of defects whose elastic properties will increase the richness of their rheology.

Therefore this paper aims to investigate the rheology of cholesteric BPs by numerically solving the hydrodynamic equations of motion for BPI, BPII and, for comparison, a simplified metastable network which we will term double twist (DT). We consider a Poiseuille flow field. Under gentle forcing we find that the viscosity is consistently higher than that of an equivalent isotropic phase but, as the forcing is increased, there is strong shear thinning corresponding to the progressive rupture of the network. As the disclination network starts to flow the periodicity of the blue phase gives rise to an oscillating state which would leave a measurable signature in the time-dependent stress curve. Elastic networks are of interest in many contexts and we compare our results to very recent studies of the rheological properties of disclinations in colloidal suspensions in liquid crystals [9].

The equilibrium properties of BPs are described by a Landau–de Gennes free energy density, \( F \), written in terms of a tensor order parameter \( Q_{\alpha\beta} \) [1,3]. This comprises a bulk term

\[
f_b = A_0 \left( 1 - \frac{\gamma}{3} \right) Q_{\alpha\beta}^2 - A_0 \gamma Q_{\alpha\beta} Q_{\beta\zeta} Q_{\zeta\alpha} + \frac{A_0 \gamma}{4} (Q_{\alpha\beta}^2)^2
\]

and a distortion term [3]

\[
f_d = K \left( \partial_{\beta} Q_{\alpha\beta} \right)^2 + K \left( \epsilon_{\alpha\zeta\delta} \partial_{\zeta} Q_{\beta\beta} + 2 p Q_{\alpha\beta} \right)^2
\]

where \( K \) is the elastic constant and \( p \equiv 2\pi/\varrho_0 \) is the pitch of the cholesteric. \( A_0 \) is a constant, \( \gamma \) controls the magnitude of order and \( \epsilon_{\alpha\zeta\delta} \) is the Levi-Civita tensor.

The time evolution of the system is governed by the equation of motion

\[
D_t Q_{\alpha\beta} = \Gamma H_{\alpha\beta}
\]

where \( \Gamma \) is a collective rotational diffusion constant and \( D_t \) is the material derivative for rod-like molecules [10]. The molecular field \( H_{\alpha\beta} \) is explicitly given by

\[
\]
\[ H_{\alpha\beta} = -A_0(1 - \gamma/3 + 4Kq_0^2)Q_{\alpha\beta} + A_0\gamma(Q_{\alpha\gamma}Q_{\gamma\beta} - \delta_{\alpha\beta}Q_{\gamma\delta}^2/3) - A_0\gamma Q_{\gamma\delta}^2 Q_{\alpha\beta} - 2Kq_0(\epsilon_{\alpha\gamma}\delta_{\beta\delta}Q_{\delta\beta} - \epsilon_{\beta\gamma}\delta_{\alpha\delta}Q_{\delta\gamma}) + 4/3Kq_0\delta_{\alpha\beta}Q_{\gamma\delta}\delta_{\gamma\delta}Q_{\delta\gamma} + KQ_{\gamma\delta}^2 Q_{\alpha\beta} - 4\frac{K}{\rho}\frac{\partial\bar{v}}{\partial\bar{p}} Q_{\alpha\beta} \] (4)

The fluid velocity, \(\bar{u}\), obeys the usual continuity equation and the Navier-Stokes equation

\[ \rho(\partial_t + \bar{u}_\beta\partial_{\bar{u}})u_\alpha = \partial_\alpha(\Pi_{\alpha\beta} + \eta\partial_\beta(\partial_\alpha u_\beta + \partial_\beta u_\alpha) + (1 - 3\partial_\alpha\partial_\beta)\partial_\gamma u_\gamma,  \] (5)

The stress tensor \(\Pi_{\alpha\beta}\) is

\[ \Pi_{\alpha\beta} = -P_0\delta_{\alpha\beta} + 2\xi(Q_{\alpha\beta} + \frac{1}{3}\delta_{\alpha\beta})Q_{\gamma\gamma}H_{\gamma\gamma} - \xi H_{\alpha\gamma}(Q_{\gamma\beta} + \frac{1}{3}\delta_{\gamma\beta}) - \xi(Q_{\alpha\gamma} + \frac{1}{3}\delta_{\alpha\gamma})H_{\gamma\beta} \] (6)

\[ \text{where } \rho \text{ is the fluid density, } \eta \text{ is an isotropic viscosity and } \xi \text{ is related to the aspect ratio of the molecules. } P_0 \text{ is a constant in the simulations reported here. Note that the order parameter field affects the dynamics of the flow field through the stress tensor. This is the backflow coupling. Details of the equation of motion can be found in [8].} \]

To solve these equations we use a three dimensional lattice Boltzmann algorithm [11,12].

In the first column of Fig. 1 we show the equilibrium configuration of the three different BPs we shall consider. The cylinders are disclination lines between regions of double twist which are repeated periodically to build up the blue phase structure. Within the tubes the order parameter attains a deep local minimum, whose value is slightly flow-dependent. Figs. IB and IC are the accepted structures of BPI and BPII. The configuration shown in IA, is the DT structure. It is the simplest disclination line network that can be constructed with double twist, i.e. a periodic array of doubly twisted cylinders with axes along z. DT is metastable and has not been observed. However its simplicity makes it a useful tool to help understand the rheology of the other blue phases. Local regions of the DT configuration are likely to be found during a temperature quench while a third observed blue phase BPIII is thought to comprise a gas of doubly twisted cylinders like the ones building up the DT network [1,3].

Each structure was obtained by relaxing \( F \) to its minimum by solving Eq. 3 numerically. Periodic boundary conditions were used and initial configurations were set according to the approximate solutions in [3]. For more details and an equilibrium phase diagram see [13].

To study the rheology of BPs, we sandwiched them between two plates a distance \( L = p/2 \) apart along the z-axis. (Qualitatively similar results are obtained with larger \( L \).) Poiseuille flow along \( y \) was imposed by a pressure gradient \( \Delta P \) together with no-slip conditions for the velocity at the plates. We quantify the strength of the forcing via the dimensionless parameter \( \tilde{F} = \frac{\Delta P}{\rho c^2} \), where \( c \) is the sound velocity. At the boundaries we assume that the director profile corresponding to the BP structures does not change under flow, that is the disclinations are fixed at the boundaries. Physically, this is likely since small surface irregularities should pin the equilibrium director profile. A similar situation occurs in permeation [8], where fixed boundary conditions give results closer to the experiments.

Fig. 2 summarises the dependence of the apparent viscosity, \( \eta_{app} \), on the strength of the forcing and compares the blue phase behaviour to that of a corresponding Newtonian isotropic fluid. \( \eta_{app} \) is found by computing the flux of \( \bar{u} \) through a plane perpendicular to y and by comparing it with that of a Newtonian fluid in the same geometry and under the same body force. For small forcing, \( \tilde{F} \lesssim 10^{-2} \), \( \eta_{app} \) increases by a factor of \( \sim 4 \) times over that of the isotropic fluid. All three phases reach a stationary state in which the networks of line defects is deformed by the flow, as shown in column II of Fig. 1. The deformations consist of first bending, and then twisting, the disclination line network without changing its topology. In BP I in particular we observe that the disclinations twist either separately or in pairs of different handedness, as predicted in [1,2].

Backflow is significant and acts to decrease the bending and twisting of the network lines. This can be clearly seen by comparing the results of the full simulations in column II in Fig. 1, to those in column III, obtained in calculations in which backflow was neglected and the order parameter was simply advected by the flow.

The primary flow is symmetric to a good approximation for BPI and BPII but displays an asymmetry for DT. This is due to the different orientation of the disclination loop with respect to the flow near the top and bottom plates, which arises as the two disclinations twist. In the case of BPI and BPII the disclinations are more numerous, so that any asymmetry is washed out by averaging. In the steady state we find non-negligible secondary flows along both the \( x \) and \( z \) directions.

For small forcing the viscosities of BPI and BPII do not vary greatly. The DT phase, however, shows shear thickening. This is because of a spectacular change in the defect structure in which each line of 2\( \pi \) disclinations (defects of topological strength 1) characterising the DT structure opens up to a disclination ring (defect of topological strength 1/2). The ring then bends and twists on itself as the flow increases. A similar transition, driven by applying an external magnetic field, has been studied in a system of colloids dispersed in a nematic [14].

We now consider what happens as \( \tilde{F} \) is increased. Fig. 2 gives evidence of significant shear thinning for all three structures considered. The disclination network is destroyed by the flow and the structure becomes that of a standard nematic subject to a Poiseuille flow [1,2].

The high and low viscosity behaviours are separated by a crossover region in which the viscosity decreases
rapidly. This corresponds to an intriguing network dynamics, shown, for the doubly twisted structure, in Fig. 3. The disclination lines in a given cell break up and part of the defect structure moves with the flow into the neighbouring cell. It then re-forms the network by attaching to the portions of the defect left behind. The process repeats and hence there is a continuous process of destruction and rebuilding of the textures. The dynamics can be monitored through the time dependence of the free energy and the stress tensor of the system, both of which oscillate (Fig. 4), at least up to the 1 ms time scale reached in the simulations.

Early experiments have reported high viscosities in BPs [15]. It is also interesting to compare our results with those obtained very recently in Refs. [9] for a disclination line network obtained by dispersing spherical colloidal particles in a cholesteric liquid crystal. These authors measured the apparent viscosity versus shear rate and found a shear thinning behaviour that closely resembles that shown in Fig. 2. Finally we comment on the rheology of cholesterics. Here there is an even higher viscosity at low shear rate, now because the director field (rather than a disclination network) does not want to distort. In both systems there is, with fixed boundary conditions, strong shear thinning at higher shear rates [8]. In cholesterics when flow is perpendicular to the helix a nematic-like viscosity is observed [16].

In conclusion, we have reported the first numerical investigation of the rheology of blue phases in cholesterics. We applied Poiseuille flow starting from three topologically different equilibrium defect networks: two correspond to the accepted structures of BPI and BPII, and the other is a periodic array of doubly twisted cylinders. Under small forcing, the network opposes the flow giving rise to a significant increase in the apparent viscosity \( \eta_{\text{app}} \). Shear forces cause the disclinations to bend and twist. In the case of the doubly twisted structure we observe a flow-induced unzipping of the array of lines into a network of loops which causes shear thickening at small forcing. As the forcing is increased, we find clear evidence of shear thinning. Similar behaviour is seen in rheological experiments on colloidal suspensions in liquid crystals. In the crossover region, as \( \eta_{\text{app}} \) drops, we also predict a novel oscillatory regime which results from the interplay of the periodicity of the blue phases with the imposed flow. Here the network continuously breaks and re-forms as portions of the disclinations in the centre of the channel move to neighbouring cells and relink with the parts of the network left behind by the flow.

We have considered a director field pinned on the boundaries at \( z = 0, p/2 \). Experience with permeative flows [8] suggests that if free boundary conditions are used there should be no increase in \( \eta_{\text{app}} \), although the rest of the phenomenology should be preserved.

Our algorithm should be applicable to study the rheology of other 3-dimensional disclination networks in liquid crystals or in liquid crystalline gels, such as those stabilised by colloids [9], emulsions or polymers. An interesting related problem is the switching in BP devices [7].

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FIG. 3. Time evolution of a DT structure under Poiseuille flow, for $F = 0.023$, for which the system is in the crossover regime, see Fig. 2. $2 \times 2 \times 1$ structural periods are shown and distinguished by different colours (the actual structure is infinite along both $x$ and $y$). The disclinations first bend (frame 2) and then unzip (3). The unzipped structure near $z = L/2$ is then extended in the direction of the flow (4) and merges with its neighbour along $y$ (5). There is some relaxation (6) and then the centres of the disclination are again stretched by the flow field (7,8) until the structure (9) resembles (4) and the process repeats. Parameters are as in Fig. 1. The times corresponding to each of the frame are also shown.

FIG. 4. Averaged stress (solid line) and free energy density (dashed line) corresponding to the dynamic evolution shown in Fig. 3. Oscillations are apparent in both curves.

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