Response of shear in bulk orientations of charged DNA rods: Taylor- and gradient-banding

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Abstract
Shear-induced instabilities leading to various kinds of inhomogeneous flow profiles play an important role in the processing of complex fluids, ranging from polymeric materials to various types of biological systems. In previously studied systems, either Taylor banding, or gradient banding, or fracture is observed. In the present work we study a system for which all instabilities occur in orientation textures (OTs), and where Taylor banding occurs simultaneously with gradient banding. The system here consists of crowded suspensions of long and thin DNA-based rods (at a low ionic strength of 0.16 mM salt), where the applied shear rate is systematically varied, for concentrations well below and above the glass-transition concentration (12.4 mg ml$^{-1}$). To simultaneously measure the velocity profile along the gradient direction, in fracture and gradient banding, the optical cell is placed in a specially designed heterodyne light scattering set up, where the scattering volume can be scanned across the cell gap. The results confirm that Taylor bands and gradient banding occur in the concentration of DNA rods and applied shear-rates (35–80 s$^{-1}$). Taylor bands clearly show the flow access in vorticity-direction, while the gradient banding is rearranged as thick rolling flows of OTs, at the middle shear-rate (50 s$^{-1}$). The observations can be then useful to facilitate other biological complex fluids and the glass-forming liquids.

1. Introduction

Complex fluids under simple shear flow exhibit several types shear-induced instabilities. A quite generic instability is the so-called gradient-banding instability, where a linear flow profile is unstable and transits to an inhomogeneous profile consisting of regions (the ‘bands’) within which the shear rate is constant, but differ between the bands. This instability has been observed for the first time in extrusion processing of polyethylene in the 1960s, leading to a sudden increase of the polymer throughput on increasing the extrusion rate (the so-called ‘spurt effect’) [1]. In an early theory on this phenomenon, the existence of interfaces between the bands is assumed on forehand [2]. Up to now, it is well understood that this instability is due to strong shear-thinning [3, 4]. Furthermore, the existence of an interface arises naturally when non-local stresses are accounted for in the constitutive relation [5, 6]. Another type of shear-induced flow instability leading to banded flow profiles has been recently discussed in colloidal systems, and is referred to as the ‘stress concentration coupling (SCC)-instability’ [7, 8], which occurs in systems with a yield stress on lowering the applied shear rate. The SCC-instability originates from shear-gradient induced mass transport. Two other types of inhomogeneous flows consist of regularly stacked regions along the vorticity direction which are alternatingly in internal rolling motion: vorticity banding and Taylor banding. Vorticity banding is most probably the result of an elastic instability [5, 9–11], while Taylor banding is due to centrifugal forces acting on fluid elements. Inhomogeneous flow profiles can be employed in the construction of micro-sensors [12, 13].
profiles due to gradient- and vorticity banding, plug flow and fraction has been observed, primarily in polymeric systems. Plug-flow velocity profiles consist of regions where the velocity is independent of position. In a fractured flow profile on the contrary, the flow velocity changes quasi-discontinuously with position at specific positions.

There is no work reported yet, where all the above-mentioned different types of inhomogeneous flow profiles are found in the same system, in dependence of concentration, with transitions between the profiles on changing the applied shear rate, and where the simultaneous coexistence of such velocity profiles has been observed. Experiments of this sort on the same system studied in the present work in [14] are limited to the rod glass state. In addition to the characterization of the flow profiles, in the present work the morphology of the system will be addressed, in equilibrium, under flow conditions, as well as the time dependence after cessation of the flow.

The equilibrium phase diagram of suspensions of fd-virus particles in the concentration-versus-ionic strength plane at low ionic strengths (below the mM-range) is discussed in [15], together with the various OTs. The primary interest arises then the difference between the presence of a shear and the absence (as the equilibrium state). On decreasing the ionic strength, the effective thickness of DNA fd-rods increases due to longer-ranged electrostatic interactions [16]. The location of an I-N transition is thereby affected by the ionic strength: both I-N binodals and spinodals shift towards lower DNA rod concentrations(fd), but also the chiral-nematic, chiral-mesophases, and glass are observed at a lower ionic strength. The equilibrium phase diagram of low ionic strengths can be found in [15].

The phase diagram for a high ionic strength (20 mM Tris/HCl buffer) is given in figure 1(a), while for a low ionic strength (0.16 mM Tris/HCl buffer) the phase diagram is shown in figure 1(b). For the low ionic strength, besides transitions between nematic and chiral-nematic structures, a glass is found at sufficiently high fd-concentrations (12.4 mg ml$^{-1}$) [17, 18]. The transition from nematic to chiral-nematic occurs on increasing the fd-concentration, where the distances between the chiral-structured cores of the fd-virus particles is diminished, and hence promoting chiral microstructural ordering. Various texture morphologies are observed in the full nematic state (see figure 1(c)). In the glass state, both the texture dynamics and the fd-particle dynamics within the domains are kinetically arrested [17, 18]. The glass state results from strong and long-ranged electrostatic inter-particle interactions [19]. The charge density of the fd-virus particles is significantly reduced by condensed ions. Since the number of condensed ions is highly susceptible to external fields, the stability of a nematic state can be reduced by applying an external electric field, leading to dynamical states under the influence of AC electric fields where nematic domains persistently melt and reform [20–22]. In this work, few representative
rod-concentrations are chosen for a low ionic strength (0.16 mM Tris/HCl buffer), to explore the deformation of orientation textures (OTs) that are formed the chiral mesophases of x-pattern, helical domains, below the glass concentration. Thus, the reason for a nematic to chiral-nematic transition, in figure 1(b), is understood by higher packing fractions of thicker electric double layers, in the ionic strength of 0.16 mM, in figure 1(c), leading to chiral mesophases OTs. Details of the equilibrium phase diagram of other ionic strengths can be found in [15]. Here, the main interest is then to show how these DNA rod glasses exhibit to the simple shear flow, from their initial caging, as well the chiral-nematic domains. This suggests then how the system responses to shear in orientations of charged DNA-rod that are self-assembled in the equilibrium, due to larger dissociation of condensed ions in bulk. The connection of the stress of homogeneously sheared systems and microstructural orders of the colloidal rods has been of interest for many decades. More recently, shear-induced instabilities leading to non-uniform flow profiles in colloidal systems have been addressed in great detail, as discussed above. How macroscopic orientational domains of colloidal systems lead to flow instabilities composing Taylor and gradient-banding as non-uniform flow profiles is not yet fully understood. The present work explores such cases, and addresses how the OTs connect to flow instabilities and the reorientation in flow profiles, to be extended existing work to quite complex, macroscopically inhomogeneous systems.

2. Sample and experimental methods: in-situ imaging heterodyne laser Doppler light scattering and image-time correlation

Experiments have been conducted for concentrations where the system is above the nematic (larger than 3.4 mg ml⁻¹), both below and above the concentration where the system forms a glass state (at 12.4 mg ml⁻¹). The samples is chosen the suspensions of charged DNA-rod (the bacteriophage fd), collected from an E-coli, for a good colloidal model system, consisting of a double-stranded DNA chain with a length of L = 880 nm, a diameter of D = 6.8 nm, shown by transmission electron micrographs [23–25]. The DNA strand is covered with 2700 fd-coat proteins, resulting in a persistence length of about 2500 nm, and a bare charge of a single fd-virus particle, varying from 6000–9600 (negative) elementary charges, depending on the pH [26]. Suspensions of fd-viruses exhibit to variable isotropic-nematic (I-N) transition in concentration [19], depending on the ionic strengths, with the effective aspect ratio, ranging from 33 to 133, at a low ionic strength of 0.16 mM, and 20 mM Tris/HCl buffer solution, respectively. It turned out that the nematic-to-chiral nematic transition occurs at sub-mM ionic strengths (below 1 mM), as well the structural glass at a unique location in the equilibrium phase diagram at higher DNA-rod concentrations (above 12.4 mg ml⁻¹) for an ionic strength of 0.16 mM [15, 17, 18]. Thus, it is an interest to exploit the shear response for the suspension of charged DNA rods at the low ionic strength of 0.16 mM salt for increasing rod-concentrations. It has also shown for shear-induced deformations of OTs in suspensions of low ionic strength DNA-rod in both real- and Fourier transform of polarized optical morphologies. In addition, the corresponding flow velocity profiles affected by the shear flow, are extensively discussed in [14].

The experimental setup of an in situ imaging heterodyne laser Doppler velocimetry light scattering (HDLS) is shown in figure 2: polarized images are captured by CCD camera in the flow-vorticity plane (see figure 2(a)), while the velocity flow profiles are measured in the top-view of gradient-flow direction (in figure 2(b)). The laser Doppler heterodyne light scattering is set on the flow-gradient direction plane, as the horizontal layout of figure 2(b), with a cylindrical shear-cell.

Two separate setups are synced in the measurement: (i) For the detection of direct imaging orientation textures (OTs), a CCD camera is set in an orthogonal direction to the laser beam, to capture the orientation texture under polarized light (between the polarizer P and analyzer A), at larger field of images (30–40 mm) through out cylindrical optical quartz cells. The crossed polarizers are placed in front and the back side of quartz optical shear cell. The total sample volume is about 6.0 ml in the gap width of 1 mm. Here, a telescopic lens is inserted to detect larger field of view, in front of CCD camera, under shear flow-vorticity plane. This is particularly useful, since the macroscopic OTs are quite large as 100–300 μm in size, through the transparent cylindrical Couette shear cell (46–48 mm diameter), which also imaging directly shear-induced deformations of OTs in the images while the flow profiles are measured. The images are shown as thought-out the shear-cell, such that the front view is slightly less focused than the backside of an image. The typical view of detection in polarized images for measurements are done with a region of interest (ROI) pixels sizes of 1292 X 968 in the real dimension of 35 × 50 mm², while the field of views for data analysis are variable.

(ii) For the detection of velocity profiles, laser Doppler velocimetry is combined with heterodyne light scattering. The laser beam alignment are aligned as follows such that an incoming laser beam is from left to the right detector side. The angle between two laser beams is 25°, so that the scattering wave vector is equal to 2.8 × 10⁷ m⁻¹. The shear cell is immersed in an index matching silicon oil bath which allows also temperature control of the sample. The cell consists of two concentric cylinders with 46–48 mm diameter where in this case
only the inner cylinder was driven by a DC servomotor and a system of belts and gear wheels to set the imposed shear rate in the range between $0.02 \text{s}^{-1}$ and $150 \text{s}^{-1}$. The lens is mounted on a nano-mover in order to sweep the focal point through the gap: the lens $L_2$ (in figure 2(b)) focuses the scattered light of the two beams onto a PMT which is connected to a correlator with a linear time spacing (see more details in [14]). Velocity profiles are measured using a home-build setup based on two incident beam laser Doppler velocimetry [27], whose details are as follows: laser is JDS Uniphase 1145P, with a power of 21 mW, and the wavelength 632.8 nm. The scattered light from the small volume $(20 \times 100 \mu m)$ where the beams are crossed, modulated from different Doppler shifts of the moving sample. The position is varied in the 1 mm gap of a transparent Couette shear cell by shifting lens $L_1$ parallel to the beams by means of a Melles Griot Nanomotion II system. The modulated light can be detected using a fiber and an ALV SO-SIPD photomultiplier assembly (PMT) in either forward or backscatter direction. The digital output of the PMT is processed by an ALV ‘FastCorr’ correlator with linear sampling time (75 ns to 400 ns) or for slow signals a homemade software. To extract the Doppler frequency from the decaying cosine correlation functions, a model function is fitted with the typical heterodyne Doppler light scattering intensity autocorrelation functions (seen in figure 2(c)), which is addressed in subsection 5.1.

3. Concentration- and shear-rate dependent 3D orientational textures in stationary flows

The appearance of a chiral-nematic phase, above the upper I-N binodal concentration, is uniquely observed for the equilibrium phase behaviors of charged DNA rods at low ionic strengths [15]. This is resultant of a delicate interplay between twist-elastic contributions and the dissociation of condensed ions among charged fd-rods. It
is not yet known whether a chiral core-structure of the single DNA-rods [28] has the origin of twist elastic energy for the collective DNA-rods, ‘restored’ effectively as the existing slow microscopic dynamics of interacting DNA-rods [29]. By increasing the DNA-rod concentration, structural glass transition is confirmed at a concentration (12.4 mg ml$^{-1}$ for the low ionic strength of a 0.16 mM salt). Below the glass-transition concentration, different OTs are equilibrated (see in figure 1(c): chiral-nematic, X-pattern, and helical domains) for a long-waiting time (80–100 h) [17, 18]. Special attention is paid to the shear-induced deformation of orientational textures. The orientational textures in the absence of shear flow vary from chiral-nematic, X-pattern, and helical domains, depending on concentration. The aim of this work is to provide the responses of simple shear flow for these equilibrated OTs under the stationary state, as well with their corresponding flow velocity profiles.

3.1. Low-concentration responses of chiral-nematic textures: stretching of orientation textures (OTs) and flow-realigning of charged DNA-rods

At above the I-N binodal (coexistent) concentration, orientation textures (OTs) of a chiral-nematic N$^*$-phase (3.7 mg ml$^{-1}$), are also observed inside the shear-cell for a cylindrical bulk shear geometry. Before applying the shear flow, the equilibration is accessed, shown in time-lapsed morphologies, collected by a CCD camera, in figure 3(a). Also the 3D intensity profiles and surface isolines are in figure 7(a), from a high shear-rate (150 s$^{-1}$) to a zero-shear-rate, at longer waiting time (8–16 h) after stopping the shear-rate. This is interesting such that the same orientation texture is equilibrated in different shear geometry (of the planar cuvette with a same thickness 1 mm), suggesting that chiral-nematic phase is a true bulk equilibrium state [15, 17, 18], disregard of shear geometry. As soon as the shear is stopped, vertically aligned nematic droplets form (within 10 s) and grow up to 8 hours, but turning to the chiral-nematic domains. This can be seen clearly, in Movie A available at stacks.iop.org/JPCO/5/045011/mmedia, as the bulk equilibrated chiral-nematic textures, checked as sustaining, in a shear cell, at least 20 h. Figure 3(b) shows the corresponding Fourier transform (FT) images of temporal changes in the deformations in 3D flow-vorticity-gradient axes. Clearly the strong elongation in FT occurs towards the flow direction right after turning off the shear, but it reorients to the vorticity-gradient directions at longer-time resting in bulk.

As the shear-response of OTs, for the low DNA-rod concentration (above the I-N binodal of 3.7 mg ml$^{-1}$), different shear-quenching are performed for (chiral) nematic droplets: (i) At a low-shear rate quenching (from a high shear-rate of 150 s$^{-1}$ to a shear rate of 0.25 s$^{-1}$), the stretching of a nematic droplet is shown to the flow direction, within 1 min, mostly the nematic droplets are stretched (see the figure 4(a)). (ii) the zero-shear quenching (from a high-shear rate of 150 s$^{-1}$ to a zero-shear quench), the flow-realigning effect is shown, in a minute after, followed by the homogenization of OTs towards the orthogonal to flow and grows in the vorticity-direction, in figure 4(b). Further visualization of such characteristic flow responses are demonstrated in the
comparison of 3D intensity profiles and surface isolines, in figure 7: the difference of shear-deformations for OTs shear-quenching is clearly depicted, where the low shear-quench is shown to be aligned in the flow direction (on the left), while the zero-quench is to the vorticity direction (on the right), respectively.

Thus, at low shear rates for low DNA rod-concentration (above the I-N transition), the local elasticity seems to play a role. In principle, such shear-induced texture elasticity can be estimated by the Ericksen number as the measure for an elastic force (against to a viscous force) in the texture relaxation. Ericksen number can be estimated by the ratio of shear-viscosity versus elasticity, as well with the applied shear-rate and the microstructures, as $Er$. The viscosity of DNA-rod concentrations are about 100–300 mPa s for the 7-19 mg ml$^{-1}$ for the nematic droplet sizes of 100–300 μm, with the texture relaxation time is about 1s. Then the Ericksen number is similar to the applied shear-rates as the order of $Er = 30 – 100$, taken for the similar range of an elasticity (as $K = 10^{-6}$ dyn) in polymeric liquid crystalline materials. These elastic forces are then originated by small nematic molecules and domains (or boundaries), likewise in the sheared polymeric nematics. It then originates from the stress-bearing structure rather than at a single domain level, thus flow responses of sufficiently low concentrations of charged DNA rods (3.4 ± 0.5 mg ml$^{-1}$) are shown to be similar with other liquid-crystalline (polymeric) materials.
3.2. A phase map of orientation textures (OTs) and flow-realigning of charged DNA-rods

The concentration- and shear rate-dependent orientation textures (OTs) under a steady shear flow are provided by a brief phase map of stationary-state for various DNA-rod concentration, at a given low ionic strength (0.16 mM Tris/HCl buffer). Here, the red vertical line indicates the glass transition concentration (12.4 mg ml$^{-1}$), established in [17, 18]. The phase-map of figure 5 shows characteristically different shear-responses, depending on the DNA-rod concentration, in the steady-state flow: (i) Far below the rod-glass, just above the isotropic-nematic coexistence concentration (3.7 mg ml$^{-1}$), as shown in previous subsection, both the stretching and flow-realigning of nematic droplets are observed in the vorticity (as Taylor bands) and gradient direction at higher rod-concentrations. Rectangular framed images are chosen for the supplementary data of 7.4 mg ml$^{-1}$, 11.1 mg ml$^{-1}$, and 14.8 mg ml$^{-1}$, in Movie B, C, and Movie D, where the thickness of rolling flow increases from 1 mm to 5 mm, and 10 mm, respectively. Here, the partial images are reproduced from [14].

**Figure 5.** The phase map of orientation textures (OTs) in the stationary state flow as a function of DNA rod concentration and shear-rate: Polarized morphologies are shown for the fd-concentration of 3.7 mg ml$^{-1}$, 7.4 mg ml$^{-1}$, 11.1 mg ml$^{-1}$, and 14.8 mg ml$^{-1}$ in the ionic strength of 0.16 mM Tris/HCl buffer. Bottom images are the low shear-rate induced homogeneous OTs, mimicking the access of equilibrium phases. The red vertical line indicates the glass transition concentration (12.4 mg ml$^{-1}$). Above the shear-rate of 50 s$^{-1}$, visibly rolling flows are present in the vorticity (as Taylor bands) and gradient direction at higher rod-concentrations. Rectangular framed images are chosen for the supplementary data of 7.4 mg ml$^{-1}$, 11.1 mg ml$^{-1}$, and 14.8 mg ml$^{-1}$, in Movie B, C, and Movie D, where the thickness of rolling flow increases from 1 mm to 5 mm, and 10 mm, respectively. Here, the partial images are reproduced from [14].
rate of 35–75 s\(^{-1}\)) and the linear-velocity profiles (at higher-shear rate of 100–150 s\(^{-1}\)) responses, which are discussed in later section 5.

Also, depending on the shear-rate, notably 'distinguishable' patterns of the OTs are induced for different DNA rod-concentration: (i) At low shear-rate (10 s\(^{-1}\)) in the phase map (figure 5), the homogenization of microstructures are observed by varying flow birefringence, under the polarized light, shown in the bottom images of figure 5. Here, the colors indicate different optical path length as the sum of optical retardance in the sample, through out the cylindrical shear-cell, under polarized light transmission. For the convenience, the blue and yellow indicates that the collective of DNA-rods are aligned to the sample, through out the cylindrical shear-cell, under polarized light transmission. To demonstrate the averaged preferred orientations of the OTs, Fourier transform (FT) images of concentration-dependent stationary state orientation textures are shown for the comparison of shear-rates (10 s\(^{-1}\) and 50 s\(^{-1}\)) in figure 6, where most pronounced shear-deformation is shown in the intermediate rod-concentration of 7.4 mg ml\(^{-1}\) in the shear rate of 50 s\(^{-1}\), showing the largest magnitude in FT spacing order in the vorticity direction, as a Taylor banding. Here, the FT is taken in the field of view as 7 mm in real space.

Further visualizations of OTs are presented in figure 7, in the 3D intensity profiles and the surface isolines, for depicting the image itself that contains the information in bulk flow (in the shear-gradient directions), while the HDLS signals are scanned few times through the gap width (1 mm) with a 20 μm space resolution. In figure 7(a), the time-lapsed morphologies 3D intensity profiles (left) and the surface isolines (right) of the equilibrated chiral-nematic OTs (at a low DNA-rod concentration 3.7 mg ml\(^{-1}\)). But they are completely deformed, when the shear is applied (see the top in figure 2(b)). However, increasing the rod-concentrations, rolling flows are shown as pronounced in the vorticity-direction (figure 7(b)), which also addressed in the following sections for varying DNA-rod concentrations and shear rates in the plane of flow-vorticity, followed by the flow velocity profiles in the plane of vorticity-gradient.

3.3. Intermediate DNA-rod concentration: Taylor-banding, from the shear-induced X-pattern and helical-domains

By an increase of the DNA-rod concentration, at intermediate concentrations, below the glass transition, the 'X-pattern' (or a Moire pattern) is observed in the shear-cell, as the orientation texture at a low shear-rate (10 s\(^{-1}\)), in figure 8. This is quite intriguing that the X-pattern is also shown as the stable equilibrium phase at this concentration \(^{[15]}\). This may indicate the low shear-rate induced homogenization could access the equilibrium phases. The Moire pattern is also reported earlier for the twist of local director (of DNA-strands) with a variable
pitch axis (see figure 5 in [35]). Although our interest is deviated from such molecular arrangements, the shear-induced collected alignments of charged DNA-rods are shown for the possible relations in the formation of Taylor-bands in the steady-state flow by two-alternating collective bright- and dark-bands in the vorticity-direction. By increasing the shear-rate of $50 - 100 \text{ s}^{-1}$, Taylor bands are shown in polarized OTs (in figure 8 (b)). This can be then understood by the delicate balance between stretching of shear-induced microstructures (consisting chiral-nematic and helical-domains) and the counter-rotating flow patterns in the vorticity-axis. Interestingly, more pronounced Taylor bands are developed at higher shear-rate above $50 \text{ s}^{-1}$, from the X-pattern at a low shear-rate.

A special note of this Taylor banding (in a current system) is that the stability of Taylor-bands are found only in the intermediate DNA-rod concentrations (7–10 mg ml$^{-1}$), for applied middle shear-rate of $50 - 100 \text{ s}^{-1}$, where the orientational helical domains, and the X-pattern exhibit slow dynamics, observed in the equilibrium phase in [15, 17, 18]. Then the low-shear-rate (10 s$^{-1}$) induced optical textures mimicking a X-pattern, in figure 8(b) may not be a coincidence. Taylor-banding is further enhanced by an instability from the non-uniform deformation of a flow (in the vorticity), as well by an elastic energy stored in orientational helical domains. Thus, this Taylor banding is different with the vorticity-banding due to hoop stresses, similar to the Weissenberg effect in polymer systems, found in the earlier work in [10, 11]. There, for fd-concentrations are chosen within the two-phase isotropic-nematic coexistence region, at a higher ionic strength (for a high aspect ratio, $L/D \sim 133$). The vorticity banding is found at very small shear rates (of the order of 1 s$^{-1}$) of complex fluids fd-virus, depleted by adding a high molecular weight of polymer (dextran). However, for the present case of the fd-suspensions at a low ionic strength (with a shorter aspect ratio of $L/D \sim 33$), no indication of

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure7.png}
\caption{Time-lapsed 3D intensity profiles (left columns) and the surface isolines (right columns) under the stationary flow, for few orientation textures (OTs) presented in figure 5: (a) for the equilibration of chiral-nematic orientation textures (of a DNA rod- concentration of 3.7 mg ml$^{-1}$) before applying the shear flow and (b) for different rod-concentrations (3.7 mg ml$^{-1}$, 7.4 mg ml$^{-1}$, 11.1 mg ml$^{-1}$, and 14.8 mg ml$^{-1}$) at a given middle shear-rate (50 s$^{-1}$). The 3D intensity profiles for field of view is 5 mm.}
\end{figure}
vorticity-banding is observed for such low shear rates. More importantly, the Taylor bands are accessed from the low-shear rate (or the equilibrated) X-pattern, which opens a new discussion whether the flow access in the intermediate concentration are maximized with these self-assembled charged DNA rods (partly discussed in [15]).

4. Image-time correlations of high-concentrations, near the rod-glass transition and 3D rolling-flows

To validate 3D orientational flows of DNA rods, velocity profiles are measured in the gradient-flow plane, by a novel experimental method of an in situ polarized imaging and heterodyne laser Doppler light scattering. An example of a heterodyne light scattering (HDLS) oscillating correlation function is shown, in figure 2 (c), under a steady-state shear-flow, performed for a Φd-concentration (11.1 mg ml$^{-1}$) and the shear-rate (1.0 s$^{-1}$). The dashed blue line is the fit of an oscillating correlation function (for a given shear-rate) at a location in the gradient direction. figure 9(a) shows the lever rule for the location of shear-rate, which is shifted from the rotating inner wall to the fixed outer wall, indicated as red, green, and blue points. The complete velocity profile is then obtained by repeating such measurements for the whole gap positions, with scanning 40–50 points in the gap width (1 mm).

Further increase of the DNA-rod-concentration, more vivid OTs are observed through the thick rolling flows: the polarized morphologies for three higher DNA-rod concentrations (10.5 mg ml$^{-1}$, 11.1 mg ml$^{-1}$ and 14.8 mg ml$^{-1}$) are shown for 2 below- and 1 in-the rod-glass concentration, respectively, in figure 9(c), for different steady shear-rates. At low shear rates, below 10 s$^{-1}$, only small domains (300 μm size) of orientation are observed. Around a shear-rate of 25 s$^{-1}$, locally developed 'twisted' OTs are aligned within the rolling flows, seen as the flow-aligning twist-patterns (at 25–80 s$^{-1}$). Also, vertically aligned 3D ‘rolling flow’ in the flow- and
vorticity-direction. In an increase of rod-concentration, the dimension of rolls increases with the rod-concentration, consequently, as the width from about 2 mm at low concentrations up to 10–13 mm at higher concentration, depicted in the right-side of figure 9(c). Eventually, at a very high shear-rate of 80–150 s⁻¹, the orientation texture melts and the velocity profile becomes linear (in the gradient-direction, on the right of figure 9(b)), also shown in the later section of velocity profiles.

To quantify the changes of polarized morphology of shear-rate dependent OTs, an image-time correlation [36] is used. Typical image-time correlation functions are performed for few shear-rates with two rod-concentrations, one is the below (11.1 mg ml⁻¹ in figure 10) and the other is above the rod-glass transition (14.8 mg ml⁻¹ in figure 11). The kinetics of OTs are then described by the slope of initial decay rates for the collected time-lapsed images. The brief procedures of constructing image-time correlation function is as follows: let \( I(t) \) be the instantaneous transmitted intensity detected by a given pixel of the CCD camera. From time traces recorded for all the pixels (in 2D matrices), the image-time correlation function \( C_{\nu}(t) \) is defined as,

\[
C_{\nu}(t) = \frac{\langle (I(t) - \langle I(t) \rangle)(I(0) - \langle I(0) \rangle) \rangle}{\langle (I(0) - \langle I(0) \rangle)^2 \rangle},
\]

where the brackets \( \langle \cdots \rangle \) denote averaging over the CCD-camera pixels. Each single image in a time trace is used to construct an image correlation function, for the region of interest as 300 × 300 pixels for the corresponding of physical dimensions of 5 × 5 mm². The above definition of the image-time correlation function is reminiscent of the scattered auto-intensity correlation function. Details of the principle of an image-time correlation can be found in [17, 18, 36], as well applied to other systems [37, 38]. Also the fitting of correlation function is as \( C_{\nu,fit}(t) \) \( \sim A \exp(-t/\tau) + B \), where \( A \) is the decay amplitude and \( B \) is the background. The results of image-time correlations are provided in figures 10 and 11, for two comparable DNA-rod concentrations, respectively, with corresponding OTs for few applied shear-rates. The image-time correlation reveals then the decays of OTs in the plane of vorticity-flow axes (via time-lapsed images in CCD-camera), while the banded flows are also measured in the gradient-flow direction (via Heterodyne Light scattering laser Doppler velocimetry, discussed in a following section). Simple sketches of rod-alignments are depicted for the glass concentration, in an increase of shear-rate, corresponding morphologies, in figure 11(b), where DNA-rods are equally aligned and dispersed in both flow and vorticity directions. Above a threshold shear-rate (25 s⁻¹), corresponding morphologies of OTs are provided on the right side of figure 11(c), coercing to the neighboring distance at high shear-rates (see the most right side of figure 11(b)). At the higher shear-rate of 35–50 s⁻¹, preferred direction of the vorticity is detected in 3D intensity profiles, with much broader 3D rolling flows in the glass concentration (14.8 mg ml⁻¹ in figure 11), compared to the below rod-glass concentration (11.1 mg ml⁻¹ in figure 10).
The major interest of image-time correlations are two folds: (i) The first is a decay time that is characteristic observation in the system: here, both concentrations have shown the faster decay rates at below the threshold shear rate (\(\sim 20^{-1}\)) for local rearrangements, but it increases again at higher shear rate (see the arrows in figures 10(b) and 11(c)), due to the imposed steady shear flow. Note that the higher concentration glass-state (14.8 mg ml\(^{-1}\)) has shown even slower decay than at two higher shear-rates, which also indicates that the glass state needs a longer time to aligning the DNA-rods in the shear flow. (ii) The second is a residual background, for instance, at a given very low-steady shear-rate (3 s\(^{-1}\)), below the glass transition concentration (in figure 10), decays of OTs occur in few sec in the steady state of shear flow, followed by uniformly aligned in the flow-vorticity direction. However, in the glass concentration (in figure 11), the initial caging of OTs exists with a small yield stress [14], which also hints that slower decays are required, below threshold shear rate (\(\sim 20^{-1}\)), in figure 11(c), before the visible response occur to the plane of flow-vorticity direction. Thus, decays of OTs in the steady state of shear flow are shown in the comparison of image-time correlations, as the shear-flow residual process in the rod-concentrations in 3D rolling flow. The simple scheme of the characteristic response of a glass-state is illustrated in figure 11(b), relating to the deformability of small initial cages in the OTs (see the most left). By an increase of the shear-rate, locally assembled orientational domains are aligned to the flow (in the middle) and rearranged to form the big rolling flows in 3D flow (at the most right). It then nicely demonstrates such distinguishable non-uniform flow responses, as the consequence of the ‘breaking’ (or ‘waking’) the boundaries of OTs, near the rod-glass concentrations, under moderately low shear-rate regimes. Whether this shear localization of glassy materials [39] can be fluidized or not (at similarly high shear-rates) could be also interested to other 3D flow behaviors of the jamming concentration for micron-sized hydrogel suspensions [40].

5. Velocity flow profiles and gradient banding of higher charged DNA rod-concentrations

5.1. Statistical averaging of HDLS velocity profiles in the gradient direction

The measurements of velocity profiles in the gradient direction are done by heterodyne laser Doppler velocimetry (HDLS), earlier mentioned in section 2. The fitting procedures of the oscillation correlations are designed by a program to calculate statistical averaging for the Doppler frequency, angled for two crossing coherent oscillating beams as,
where $\theta_s$ is the scattering wavevector for a given refractive index $n$, wavelength of laser as $\lambda = 633$ nm. The angle $\theta_s$ must be taken in the sample medium, so that for small angles frequency, $f$ is independent of the refractive index $n$ when the outside angle is measured. The fringe spacing as, $\delta = \frac{\lambda}{2 \sin(\theta_s/2)}$, for the grating of two laser beams crossing at an angle $\theta_s$ can also be used to calculate the velocity from a Doppler frequency, $f = \frac{c \cos(\beta)}{\delta}$. The angle between the incident beams (ca. 25°) and the laser wavelength. To extract the Doppler frequency from the decaying cosine correlation functions, a fitting model function is used by,
where $B$ accounts for a non-zero baseline, $A$ for an amplitude different from 1 and $A_1$ for a not perfect modulation of the measured correlation function. Brownian motion (likewise in normal diffusion) shows the linear decay term with $\tau_{\text{lin}}$, while a common speed of the whole scattering volume or shear is then expected to be influence as $\tau_{\text{agf}}$. Typical performances of location-dependent velocity profiles (in gradient directions) are collected (also seen as figure 2(c)), and total measurement positions are about 40 data locations of 5–6 data points in a single position for imposed 4–5 oscillations settings. The laser beam is scanning from the outer wall to the inner wall, much rigorously imposed oscillations are shown near the inner wall, compared towards outer wall. The envelop velocity file is then obtained from these automated statistical averaging of much data analysis processes.

5.2. Gradient banding of DNA-rod glass concentrations, above the X-pattern

Extensive studies of non-uniform flows of DNA rod-glasses are shown in [14], where strong shear thinning behaviors lead to the gradient-banding in this system. There, the internal fracture (or the plug flow), gradient banding, and the linear flow profiles are addressed as the soft rod glass with the shear stress as a function of concentration up to very high glass concentrations [14]. For a concentration of 11.1 mg ml$^{-1}$, plug flow is observed with a pronounced slip up to shear rates 30 s$^{-1}$. Close to and within the glass state, on the contrary, a combination of plug flow and fracture is observed up to shear rates of 40 s$^{-1}$, without slip. For both concentrations, there is a transition to gradient banding at a shear rate of about 30–40 s$^{-1}$, and a subsequent transition to a linear flow profile at a 150 s$^{-1}$. Here, to extend a discussion, without losing an originality of the work, velocity flow profiles of 4 (representative) concentrations are shown with the shear deformations of OTs, two for below- and the other two in the glass concentrations: different flow behaviors are observed in the gradient direction, corresponding response of OTs, under the applied shear: (i) When the shear-rate is small

Figure 12. The velocity profiles (in the gradient-flow plane) for 4 representative DNA rod-concentrations, two below the glass transition (7.4 mg ml$^{-1}$ and 11.1 mg ml$^{-1}$) and the other two glass concentration (14.8 mg ml$^{-1}$ and 19.0 mg ml$^{-1}$); (a) for the ‘plug’ flows at low shear-rates (0–25 s$^{-1}$), and (b) clear banded flows, in the mid-shear rate (35–80 s$^{-1}$), as well the linear flow profiles, at even higher shear-rates (80–150 s$^{-1}$). The upper right arrows indicate a rotating inner wall against the fixed outer wall (the left blue line), where the laser beam scans the range of entire distance through the gap width 1 mm. The imposed shear-rates are indicated as the right-side arrows. (c) Shear-induced collective alignments of DNA rods are shown in both the real- and FT-images for the rod-concentrations, at a given shear-rate (75 s$^{-1}$). As an increase of rod-concentration, the systematic decrease of FT-spacing is also shown in the vorticity direction.

$$g_2(t) = B + A \exp \left\{ -\left( \frac{t}{\tau_{\text{lin}}} \right) \right\} \left\{ A_1 + \cos \left( \frac{2\pi t}{\tau_{\text{osc}}} \right) \right\},$$

(3)
(below 10–25 s\(^{-1}\)), mostly the plug flow (or internal fractures) are shown near the inner rotating walls, indicted as the short arrows in figure 12 (a). The velocity profiles are independent throughout the shear gradient gap in position. (ii) However, in an increase of the shear rate above the threshold value (35–50 s\(^{-1}\)), clear gradient-banding is observed with the slight shift of the intersection points of banded flows in the gradient direction, in figure 12(b). The wall-slips are not present at these high rod-concentrations in the inner rotating wall. At very high shear rates (100–150 s\(^{-1}\)), the linear velocity flow profiles are found, indicating that the microstructures are greatly realigned to the flow direction. These features also agree with the Fourier transform images in figure 12(c) at the intermediate shear-rate (75 s\(^{-1}\)), measured for four different concentrations, two for below- (as 7.4 mg ml\(^{-1}\) and 11.1 mg ml\(^{-1}\)) and two for above the glass transition concentration (as 14.8 mg ml\(^{-1}\) and 19.0 mg ml\(^{-1}\)), respectively.

In addition, more pronounced Fourier spacing is shown in the direction of vorticity against the flow direction, in figure 12 (c), which are seen rigorously in the real space as well. As an increase of rod-concentrations, the FT spacing is reduced due to the increase of thickness of rolling flow, except the glass concentration in equalizing both flow and vorticity directions. The most pronounced sharp linear FT peaks are obtained in the intermediate DNA rod-concentration of 7.4 mg ml\(^{-1}\), where Taylor bands are formed. Taylor banding is found at lower shear rates as compared to gradient-banding, which is attributed to the elasticity of OTs. Interestingly, this concentration is the stable X-pattern (observed in the equilibrium phase) without a shear [15]. This strongly suggest that the intermediate concentration of X-pattern carries out a unique feature for charged DNA-rod to response in shear (see the Moire-pattern at a low-shear rate of 10 s\(^{-1}\) (see figure 8(b)); Below this concentration, OTs of chiral-nematic domains melt in the flow, while above concentrations, the OTs of chiral mesophase are interconnected to the flow with ‘twisted’ manner to form thicker 3D rolling flows. Thus, the possible driving mechanism of forming the OTs are apparently different in terms of their ‘rotatory power’, driving such delicate shear response for showing Taylor bands and thick 3D Rolling flows, in terms of the rod-concentrations and shear rates.

6. Conclusion

The system of concentrated charged DNA-rod exhibits nicely in the stationary state response to both shear-rate and the concentration-dependent non-uniform bulk orientational flow profiles. The fractured ‘plug’ flow, Taylor-banding, and gradient-banding are occurred, as resulting the flow responses of OTs, originated by existing chiral-nematic domains at low ionic strengths. These non-uniform flows depend on the concentration of DNA-rod, and their different shear-responses of OTs are obtained by the in situ imaging and heterodyne laser Doppler velocimetry. The brief summary of shear-responses for rod-concentrations are as follows: (i) Far below the rod glass concentration, above the isotropic-nematic coexistence (as 3.7 mg ml\(^{-1}\)), shear-induced stretching of nematic droplets (at a low shear-rate), as well the flow-induced realignment of rods are observed differently by a shear-quenching processes. The nematic droplets melt, at even below the threshold shear rates (25–35 s\(^{-1}\)), in the shear flow (see figure 5). (ii) However, in an increase of the intermediate DNA-rod concentrations (in the range of 7.4–9.4 mg ml\(^{-1}\)), chiral mesophases of X-pattern and helical domains are formed, as well Taylor-bands appear above the threshold shear rates (35–50 s\(^{-1}\)). This is related to a delicate balance between the fluid and orientational alignments of OTs, in the vorticity-flow plane, leading to the formation of Taylor bands. These Taylor-bands are not yet fully developed to the gradient direction, checked by the velocity flow profile in the gradient-direction. Gradient-banding occurs, in the vicinity of a rod glass transition (12.4 mg ml\(^{-1}\)) and within the glass concentration, confirmed by the velocity flow profiles within the rolling flows (in the vorticity-flow planes). (iii) At higher concentrations, much thicker bulk rolling-flows are shown along entire chiral mesophase domains in the ‘twisted’ in flow-aligned directions (see the supplementary data of Movie B, C, and D).

Also, by varying the shear rates, three distinguishable velocity profiles are obtained, in an increase of shear-rate: (i) The ‘plug’ flow, at low shear-rates (below 10–25 s\(^{-1}\)), for various homogenized OTs at given DNA-rod concentrations. (ii) Gradient-banding flow, at the middle range of shear-rates (35–80 s\(^{-1}\)), higher than a threshold shear-rate (35–50 s\(^{-1}\)), and finally (iii) the linear velocity profile at the highest shear-rate (100–150 s\(^{-1}\)).

To conclude, there are few highlights for the response of shear in bulk orientations of charged DNA rods:

• The relation between the X-pattern (in the intermediate rod-concentration) and Taylor-banding reveals that a stronger elastic response of the chiral-mesophase domains in X-pattern in the vorticity direction. Thus, the response at low shear rate is mimicking the equilibrated phase, while at high shear-rate deformations is rearranged by collective dynamics of orientation textures (OTs) adapting to the vorticity-flow direction, as Taylor bands. Most pronounced Taylor banding is shown in DNA rod-suspensions in the intermediate rod-
concentration (7.4 mg ml$^{-1}$), at moderate shear-rates (50–75 s$^{-1}$), by accessing sufficient flow to the vorticity-direction. Realizing this kind of assessments are important to facilitate other types of rod-suspensions in the stable bulk phase of anisotropic charged particles.

- The gradient-banding is occurred in the concentrations of soft rod-glass. Gradient banding exhibits earlier formation in the rolling flow at lower shear rates than that of Taylor bands, in the vorticity-gradient-flow directions. They are also shown with interconnected locally ‘twisted’ OTs along the flow line, which are visualized by the image-correlations and velocity flow profiles in HDLS measurements. Also, for low shear-rates (below 25 s$^{-1}$), the frozen-in domains are deformed to rearrange, by rendering the glass brittle, and leading to a plug flow, before the gradient-banding occurs at the middle shear rate (35–85 s$^{-1}$).

- In an increase of rod-concentration, the systematic decrease of FT-spacing is also shown in the vorticity direction. When the shear flow is sufficiently strong (above 100–150 s$^{-1}$), the linear-velocity profiles are obtained for all rod-concentrations, indicating that these OTs of rod-glasses are ‘fragile’ as the soft glasses. On the contrary, for the low rod-concentration, the (chiral) nematic droplets melt even below the threshold shear rate (20–25 s$^{-1}$), while the shear deformation of OTs exists at higher rod-concentrations, above the threshold shear rate.

Finally, this work confirms that the current single system nicely demonstrates non-uniform flow bands in the gradient-direction, as well the pronounced Taylor bands, from the low-shear-rate fractures and ‘plug’ flows. The results are useful for employing 3D orientations of rods, which may incorporate the directional flow processing, originated by different self-organized microstructures in other biological complex fluids and the synthetic rod-macromolecules.

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**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

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