Smectic-like rheology and pseudo-layer compression elastic constant of a twist-bend nematic liquid crystal

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In twist-bend nematic (NTB) liquid crystals (LCs), the mean molecular orientation exhibits heliconical structure with nanoscale periodicity. On the mesoscopic scale, NTB resembles layered systems (like smectics), where the helical pitch is equivalent to “pseudo-layers” without a true mass density wave. We study rheological properties of a NTB phase and compare the results with those of an usual SmA phase. Analysing the shear response and adapting a simplified physical model for rheology of defect mediated lamellar systems we measure the pseudo-layer compression elastic constant $B_{eff}$ of NTB phase from the measurements of dynamic modulus $G^*(\omega)$. We find that $B_{eff}$ of the NTB phase is in the range of $10^8 - 10^9$ Pa and it follows a temperature dependence, $B_{eff} \sim (T_{TB} - T)^2$ as predicted by the recent coarse-grained elastic theory. Our results show that the structural rheology of NTB is strikingly similar to that of the usual smectic LCs although the temperature dependence of $B_{eff}$ is much faster than smectic LCs as predicted by the coarse-grained models.

Experimental discovery of twist-bend nematic (NTB) phase in bent-core liquid crystals has created immense interests in the scientific community [1–7] although it was theoretically predicted much before from different perspectives [8–11]. In the NTB phase, the director $\hat{n}$ (mean molecular orientation) exhibits periodic twist and bend deformations forming a conical helix and is tilted with respect to the axis $\mathbf{N}$ as shown in Fig.1(a). The typical pitch $p$ of the heliconical structure is of the order of 10nm, thus comparable to a few molecular length. Commonly, NTB phase is observed in odd-membered liquid crystal dimers in which two mesogenic units are connected through a flexible spacers [12–14]. A fascinating feature of the NTB phase is the observation of spontaneous chirality i.e., formation of both left and right handed helical domains even though the constituent molecules are achiral. This leads to several unusual physical properties of NTB phase compared to the conventional nematic phase (N) [15–21].

A few coarse-grained theories have been proposed to explain the emergence of NTB phase from the high temperature uniform nematic phase. Mayer and Dosov showed that the elastic properties of the NTB phase could be viewed in two different length scales in reference to the pitch length ($p$) [22]. When the considered length ($l$) is less than $p$ i.e., $l < p$, the elastic description is similar to that of the usual nematics. On the other hand, when $l >> p$, the elastic description is similar to regular lamellar systems such as cholesteric and smectic LCs [22]. In the latter picture, the thickness of one pitch can be considered as a pseudo-layer and the large-scale elasticity of the NTB phase could be described in terms of an effective pseudo-layer compression elastic constant $B_{eff}$ and an curvature elastic constant $K_{eff}$ and the corresponding free energy density of NTB can be expressed as [13]:

$$f_{TB}^{CG} = \frac{1}{2} B_{eff} \epsilon^2 + \frac{1}{2} K_{eff} \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2$$  \hspace{1cm} (1)

where $\epsilon$ is the pseudo-layer compression and $R_1$, $R_2$ are the curvatures. Another coarse-grained theory has been developed considering the heli-polar order and their coupling with bend distortions [20, 23]. Both the theories predicted that the temperature dependence of $B_{eff}$ is much faster than the usual SmA LCs.

There have been a very few experimental studies reporting measurements of $B_{eff}$ of NTB LCs [23–25]. For

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FIG. 1. (a) Schematic view of heliconical molecular orientation of the nematic twist-bend (NTB) phase. $p$ represent helical pitch equivalent to pseudo-layer thickness. (b) Chemical structure of CB9CB molecules. (c) Polarising optical microscope texture at 94°C in the NTB phase. $\mathbf{N}$ is the macroscopic average orientation of $\hat{n}$ over several periods.
example, Gorecka et al. have measured $B_{eff}$ of CB7CB and some chiral $N_{TB}$ LCs. They reported the values are in the range of a usual SmA LC ($10^6$–$10^7$ Pa) and varies inversely with the temperature [24]. Parsouzi et al. have reported that $B_{eff}$ is in the range of $10^3$–$10^4$Pa and it scales as $B_{eff} \sim (T_{TB} - N)^{3/2}$ [23]. Thus, several orders of magnitude difference in the reported values, measured on two different samples using two different methods and its universal temperature dependence is still an unresolved problem. In this paper we report experimental studies on the rheological properties of a $N_{TB}$ LC. We use a novel method for measuring $B_{eff}$ from the dynamic shear modulus $G^*(\omega)$. We discuss the temperature dependence of $B_{eff}$ and compare with that proposed by the coarse-grained elastic theories of the $N_{TB}$ phase.

The LC material 1,ω-bis(4-cyanobiphenyl-4’-yl) alkane (CB9CB) studied was synthesised in our laboratory. It is a cyanobiphenyl-based dimer with odd number methylene units ($n = 9$) in the flexible spacer (Fig.1(b)). It exhibits following phase transitions: I 124°C N 108°C $N_{TB}$ 84°C Cr. and enantiotropic transition with widest temperature range of $N_{TB}$ phase (22°C) among the LCs in the homologous series of CBnCB [14]. For the purpose of comparison we also studied 8CB (Octylcyano biphenyl) which shows following phase transitions: I 41°C N 34°C SmA 22°C Cr. We used a strain controlled Rheometer (MCR 501, Anton Paar) with a cone-plate measuring system having a plate diameter of 25mm and cone angle of 1° for rheological measurements. A Peltier temperature controller was attached with the bottom plate for controlling the temperature with an accuracy of 0.1°C. A hood was used to cover the measuring plates for uniformity of sample temperature. Temperature dependent viscosity was measured in cooling the sample from the isotropic phase. For measuring the dynamic shear modulus the sample was quenched from the isotropic to the $N_{TB}$ phase. A total 5g LC was synthesised and about 200mg was used for each rheological measurement. Initially the phase transitions and textures were observed using a polarising optical microscope (Olympus BX51) and a temperature controller (Mettler FP 90). A typical texture of an unaligned sample is shown in Fig.1(c). It is noticed that the textures of $N_{TB}$ are very similar to that of the focal conic textures of usual SmA LCs.

To begin with we measured shear viscosity of CB9CB as a function of temperature to identify the phase transition temperatures. Since the orientation of the director with respect to the shear direction changes with temperature we define it as effective viscosity $\eta_{eff}$. As shown in Fig.2(a) the N to $N_{TB}$ transition is identified from the rapid increase (more than two orders of magnitude) of $\eta_{eff}$ with respect to the N phase. The onset of the N-$N_{TB}$ transition (108°C) is better seen in the inset of Fig.2(a). We also measured temperature dependent $\eta_{eff}$ of 8CB LC as shown in Fig.2(a). It is evident that the magnitude and the overall temperature dependence of $\eta_{eff}$ the two samples are very similar. It possibly indicates that the pretransitional fluctuations and the resulting director dynamics across the N-$N_{TB}$ transition are similar to that of N-SmA transition [26, 27]. The large $\eta_{eff}$ of the $N_{TB}$ phase is expected to arise from the perpendicular and parallel orientations of the pseudo-layers similar to those reported in the SmA phase of 8CB LC [28]. The details of these orientations are discussed later. The flow curve of $N_{TB}$ phase has been studied and compared with that of the SmA phase of 8CB as shown in Fig.2(b). $N_{TB}$ shows both shear-thinning and yield stress similar to that of the SmA phase of 8CB LC. The shear rate dependent shear stress $\sigma_{eff}$ is fitted with the Herschel-Bulkley (HB) model:

$$\sigma_{eff} = \sigma_y + A\dot{\gamma}^n$$

where $\sigma_y$ is the yield stress, and $A$ and $n$ are constants. The fit parameters obtained are: $n = 0.6$, $\sigma_y = 2.4$ Pa, $A = 2.1$ for $N_{TB}$ and $n = 0.65$, $\sigma_y = 7.5$ Pa, $A = 2.8$ for SmA phase. The two sets of fit parameters characterising the flow curves of two phases are reasonably close, suggesting they have similar flow behaviour under shear.

FIG. 2. (a) Temperature dependent effective viscosity $\eta_{eff}$ of CB9CB (blue squares) and 8CB (black spheres) LCs. (Inset) Variation of $\partial \eta_{eff}/\partial T$ with temperature $T$ of CB9CB. Shear rate $\dot{\gamma} = 100$ s$^{-1}$. (b) Shear rate dependent effective shear stress $\sigma_{eff}$ at fixed temperatures. The solid lines are theoretical fits to Eq.(2).
FIG. 3. (a) Strain amplitude dependence of the storage $G'$ (solid symbol) and loss moduli $G''$ (open symbol) for N$_{TB}$ ($T=87^\circ$C) and 8CB ($T=26^\circ$C). Solid line is a theoretical fit to Eq.(3) (b) Temperature dependence of $G'$ (solid symbol) and loss moduli $G''$ (open symbol) at a fixed strain amplitude $\gamma = 0.1\%$. Measurements are performed at frequency $\omega = 1$ rad/sec.

As a next step, we measure the dynamic modulus $G^* = G' + iG''$. The regime of linear viscoelasticity of N$_{TB}$ is determined by performing oscillatory measurements in which the strain amplitude is varies from $\gamma = 0.01\%$ to $\gamma = 100\%$ at a fixed frequency $\omega = 1$ rad/sec. The strain amplitude dependence can be described by the empirical relation [29],

$$G'(\omega, \gamma) = \frac{G'(\omega, 0)}{1 + \gamma/\gamma_c}$$

where $\gamma_c$ is the critical strain amplitude. As shown in Fig.3(a) the critical strain amplitude $\gamma_c = 3.3\%$ and modulus $G'(1, 0) = 391$ Pa, setting the upper limit of linear viscoelastic regime. For comparing we also measured $G^*$ of the SmA phase of 8CB and shown in Fig.3(a). It is interesting to note that not only the strain dependence but also the magnitudes of the shear moduli of the N$_{TB}$ and SmA are comparable, indicating they have common structural origin. Further we measured the temperature dependence of dynamic moduli of N$_{TB}$ at a fixed shear amplitude $\gamma = 0.1\%$ and observed that in the entire N$_{TB}$ phase $G' > G''$. The shear response of the N$_{TB}$ phase can be discussed in analogy with the rheological responses of usual SmA liquid crystals [29]. Like SmA, N$_{TB}$ is solid like in one direction and liquid like in other two directions. Three simplest orientations of the pseudo-layers can be considered with respect to the shear ($\vec{v}$) and velocity gradient ($\vec{\nabla}v$) directions. When the pseudo-layers are orientated parallel to the plane defined by $\vec{v}$ and $\vec{\nabla}v$ the N$_{TB}$ phase behaves like a liquid. Similarly when the pseudo-layers are orientated parallel to the shear plane they can slide past each other easily and the N$_{TB}$ phase behaves like a liquid. When the pseudo-layers are perpendicular to the shear direction ($\vec{n}$ is parallel to $\vec{v}$) shear tends to change the pseudo-layer spacing. As a result of which N$_{TB}$ phase shows a viscoelastic solid-like behaviour consequently, $G' > G''$. Assuming the pseudo-layer spacing $p \approx 10$ nm, the estimated modulus $G' \approx kT/p^3 = 4 \times 10^3$ Pa, which is nearly one order of magnitude larger than $G'(1, 0)$ (see Fig.3(a)). It means the length scale involved is larger than the pseudo-layer spacing which could arise from the defect texture of the unaligned sample.

FIG. 4. Frequency dependence of storage $G'$ (solid symbols) and loss $G''$ (open symbols) moduli at a few representative temperatures. Solid lines are theoretical fits of Eq.(4) and Eq.(5) to $G'$ and $G''$, respectively.

The observed solid-like response of the N$_{TB}$ phase can be explained based on a simple physical model described for defect mediated cholesteric and smectic LCs. According to this model the storage modulus can be expressed as [30]

$$G'(\omega) = G_0 + \beta_1\omega^{1/2} + \beta_0\omega^2$$

The first term $G_0$ arising from the elasticity of the static defects [31]. The second term $\beta_1\omega^{1/2}$ arises from the regions of misaligned pseudo-layers in the sample [32].
last term results from the regions of the sample where the pseudo-layers are parallel to the shear direction. The proportionality constants are given by $\beta_0 \simeq \eta (\gamma_1/K)(p/4\pi)^2$ and $\beta_d = (\pi/24\sqrt{2})\sqrt{(B_{eff}\eta)}$ where $\gamma_1$ is the rotational viscosity, $\eta$ is the effective viscosity and $B_{eff}$ is the elastic compression modulus [32]. In particular, $\beta_d$ describes the response of the lamellar regions with the layer normal oriented such that strain involves layer compression. Following the similar arguments the loss modulus can be written as [30]

$$G''(\omega) = \alpha_d\omega^{1/2} + \alpha_0\omega$$  \hspace{1cm} (5)$$

where the first and the second terms arise from the misaligned parts of the samples and the Maxwell-fluid type contribution, respectively. Thus, by measuring $\beta_d$ and $\alpha_0$ from the disoriented sample, we can estimate $B_{eff}$. In order to measure these parameters at different temperatures we quenched the sample directly from the isotropic to the N$_{TB}$ phase so as to obtain mostly disoriented sample. Figure 4 shows some representative plots of $G'(\omega)$ and $G''(\omega)$ at different temperatures in the N$_{TB}$ phase. The parameters obtained by fitting Eq.(4) and Eq.(5) at different temperatures are shown in Fig.5, except $\beta_0$, which is very small ($10^{-3}$) and does not vary considerably with temperature. It is noted that $\beta_d \simeq \alpha_d$ as expected theoretically and both increases with decreasing temperature. Assuming the whole sample is in the disoriented state, the temperature dependence of $B_{eff}$ can be expressed as

$$B_{eff} = \frac{\beta_d^2}{\alpha_0} \left( \frac{24\sqrt{2}}{\pi} \right)^2$$  \hspace{1cm} (6)$$

Figure 6 shows the temperature dependence of calculated $B_{eff}$ in the entire N$_{TB}$ phase. Just below the N-N$_{TB}$ transition $B_{eff}$ is relatively smaller; $1.5 \times 10^3$Pa at $T=105^\circ$C and it increases rapidly to $2 \times 10^6$ Pa at $T=85^\circ$C. The latter number is only one order of magnitude lower that the typical layer compression modulus of SmA phase of 8CB [33]. So far experimentally $B_{eff}$ of a very few N$_{TB}$ LCs have been measured. Gorecka et al. measured temperature dependence of $B_{eff}$ of a few chiral twist bend nematic LCs including CB7CB using atomic force microscopy technique and reported that $B_{eff}$ is in the range of $10^6 - 10^7$ Pa of an ordinary SmA LCs. Using a dynamic light scattering technique, Parsouzi et al. reported that $B_{eff}$ of N$_{TB}$ phase of a LC made of multi-component mixture is in the range of $10^3 - 10^4$ Pa, which is almost three orders of magnitude smaller than the ordinary SmA LCs [23, 25]. Our experiment shows a wide variation of $B_{eff}$, covering both the ranges. Such wide variation of $B_{eff}$ could partly be attributed to the increase in the cone angle $\theta$ with decreasing temperature. Based on a crude model $B_{eff} = K_2(2n/p)^2\sin^4\theta$ [25], where $\theta$ increases with decreasing temperature [19, 34]. Considering physical parameters for CB7CB such as: $p = 10nm$ [19] and $\theta = 10^\circ$ [34] (2°C below N-N$_{TB}$ transition); $p = 8nm$ [19] and $\theta = 33^\circ$ [34] (25°C below the transition) and $K_2=3pN$, the calculated $B_{eff}$ at two far away temperatures are given by $1.1 \times 10^3$ and $1.6 \times 10^5$ Pa. The calculated $B_{eff}$ close to the transition agrees reasonably well with our experiments but it is smaller by one order of magnitude at far below the transition. This assessment suggests that mere increase in the cone angle with decreasing temperature can enhance $B_{eff}$ by almost two orders of magnitude.

The temperature dependence of $B_{eff}$ have been predicted theoretically using coarse-grained theoretical models. In analogy with SmA*, Meyer and Dosov defined smectic-like effective pseudo-layer compression and bending elastic constants ($K_{33}^N$) [22]. Assuming $K_{33}^N < 0$, they predicted $B_{eff} \sim (T_{TB}-T)^2$. Parsouzi et al. have developed another model accounting the helical polarisation field and their coupling with the bend distortion of the director [23]. Considering a small variation in the pseudo-layer spacing and resulting changes in the cone angle and polar order, the theory predicts that there are two regimes of $B_{eff}$. For temperature (T) sufficiently close to the T$_{TB}$, $B_{eff} \sim (T_{TB}-T)^3$, whereas for T sufficiently below T$_{TB}$, the theory gives $B_{eff} \sim (T_{TB}-T)^{3/2}$. Experimentally they found (in a mixture exhibiting N$_{TB}$) that within a relatively small temperature range ($\sim 6^\circ$), $B_{eff}$ scales as $(T_{TB}-T)^{3/2}$. To get an estimation of the scaling exponent of $B_{eff}$, we fit our data to the equation $B_{eff} \sim (T_{TB}-T)^{\alpha}$, with $\alpha$ as a fit parameter as shown.
in Fig.6. We obtain $\alpha = 2.0 \pm 0.1$, which is equal to the scaling exponent predicted by the “negative elasticity” model of Meyer and Dosov [22].

The temperature dependence of $G_0$ as shown in Fig.5(b) indicates that the static defects contributes to the mechanical response of the N$_{TB}$ phase similar to that of the SmA phase. Inset of Fig.5(b) shows that it scales with temperature as $G_0 \sim \chi$, where $\chi = (T_{TB} - T)/T$ is the reduced temperature. This is slightly faster than that in case of SmA ($\chi^{0.7}$)[38] as expected as in the N$_{TB}$ phase $B$ scales much faster with temperature. Two remarks are in order. Firstly, in estimating $\beta_d$ it has been assumed that the pseudo-layers are disoriented in the whole sample. This assumption is reasonable as the sample was quenched from the isotropic phase. Nevertheless if the sample is partly oriented we do not expect an order of magnitude reduction of $\beta_d$ and consequently $B_{eff}$. Following the same procedure $B_{eff}$ of 8CB is measured at a few temperatures which agrees well with that reported [33]. Secondly, the experimental results indicate that there are defects in our system whose contribution to the elasticity increases linearly with decreasing temperature. This might indicate a type-II N$_{TB}$ having twist grain boundary (TGB) like structure as predicted by the coarse-grained model [22]. However, at this point we can say that more experimental investigations are required to draw any unambiguous conclusion.

In this work we have presented rheological properties of a N$_{TB}$ LC. The structural rheology of N$_{TB}$ phase is found to be remarkably similar to that of the usual SmA phase of calamitic liquid crystals. Our measurements reveal that in spite of the absence of true mass density wave, N$_{TB}$ LCs are viscoelastic solids similar to many defect mediated lamellar systems. We found that $B_{eff}$ is relatively softer near the N-N$_{TB}$ transition but increases to three orders of magnitude more at a much faster rate than the usual SmA LCs with decreasing temperature. The temperature dependence of $B_{eff}$ agrees well with the prediction of the coarse-grained elastic theories. Thus our results provide a valuable test of the validity of the proposed theoretical models. The experiments also offer new perspectives of N$_{TB}$ LCs and open unexplored aspects of rheology of nematic LCs with nanoscale modulation of director.

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