

Flow alignment in a shearing nematic liquid crystal near a charged surface

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The flow alignment angle $\theta_{eff}(y)$ and effective rotational viscosity coefficient γ_1^{eff} are investigated for polar liquid crystals (LCs), such as 4-*n*-octyloxy-4'-cyanobiphenyl (8OCB), in the vicinity of a charged bounding surface. γ_1^{eff} and $\theta_{eff}(y)$ are calculated, for the stationary regime, in the framework of the conventional Ericksen - Leslie theory. Calculations of γ_1^{eff} , for the homeotropic alignment of 8OCB molecules, at a charged indium tin oxide-coated glass plate show an increase in γ_1^{eff} up to 7.8%.

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The flow alignment of a nematic liquid crystal (NLC) characterized by a unit vector along the optical axis (the director \mathbf{n}) is governed by a shear flow \mathbf{v} . If the director is aligned in the shear plane (x - y plane defined by the liquid crystal flow (x direction) and the velocity gradient in the y direction; z is the vorticity axis) prior to flow, the hydrodynamic torque exerted per unit volume is given by [1] $T_{vis} = \frac{1}{2}(\gamma_1 + \gamma_2 \cos 2\theta_{bulk})\dot{\gamma}$ and vanishes when the director aligns at an angle

$$\theta_{bulk} = \frac{1}{2} \cos^{-1} \left(\frac{1}{\lambda} \right), \quad (1)$$

with respect to the flow direction. Here $\lambda = -\gamma_2/\gamma_1$, γ_1 and γ_2 are the rotational viscosity coefficients (RVCs), and $\dot{\gamma}$ is the shear rate. However, in the vicinity of the bounding surface the dynamics of NLCs is also dependent on the surface potential, which penetrates the bulk nematic over a distance ξ , and gives an additional contribution to the torque balance. As a result, the flow alignment angle $\theta_{eff} = \frac{1}{2} \cos^{-1}(\lambda_{eff}^{-1})$ is also governed by the strength of the surface potential, which may include both short- and long-range contributions. Several short-range surface potentials have already been studied in the past [2–4]. For example, it was shown that a short-range substrate-particle interaction based upon the integrated Lennard-Jones form adopted in adsorption studies of *cyanobiphenyls* on graphite [5], leads to the surface energy [6]

$$f_0 = -\frac{1}{2} w_0 (\mathbf{n}_s \cdot \mathbf{n}_0)^2 = -\frac{1}{2} w_0 \cos^2(\theta_s - \theta_0), \quad (2)$$

where w_0 is the anchoring strength, θ_s and θ_0 are the polar angles (with respect to the planar normal) of a surface director \mathbf{n}_s and an easy axis direction \mathbf{n}_0 [6], respectively. The surface energy delocalizes over a thin bulk layer of thickness λ_s (of the order of 10–100 nm [2,7]) close to the substrate. Over the distance λ_s , the nematic order parameter changes from its value at the surface to that of the bulk sample. When

the solid substrate is in contact with the nematic, selective ion adsorptions take place. For instance, the positive ions are attracted by the substrate, whereas the negative ones are repelled. In this case, the surface electric field E_0 , originating from the surface charge density σ , will penetrate the bulk on the order of the Debye screening length λ_D [8], owing to ions present in the bulk of the liquid crystal (a weak electrolyte). The distance dependence of the surface electric field with bulk screening is given by [8]

$$\mathbf{E}(y) = E_0 \exp\left(-\frac{y}{\lambda_D}\right) \mathbf{j}, \quad (3)$$

where $E_0 = \sigma/\epsilon_0 \bar{\epsilon}$ is the surface electric field of the charged plane, ϵ_0 is the absolute dielectric permittivity of free space, $\bar{\epsilon} = (\epsilon_{\parallel} + 2\epsilon_{\perp})/3$ is the average dielectric permittivity, ϵ_{\parallel} and ϵ_{\perp} are the dielectric constants parallel and perpendicular to the director \mathbf{n} , respectively, \mathbf{j} is the unit vector directed away and perpendicular to the substrate, and y is the distance from the surface in the \mathbf{j} direction. In this paper we attempt to answer questions, based upon the conventional Ericksen-Leslie (EL) [9,10] theory, of how the surface charge density σ affects the flow alignment angle θ_{eff} at distances between λ_s and $6\lambda_D$ from the substrate and the dissipative processes described by the rotational viscosity in the same region. It should be pointed out that in practice it is difficult to measure dynamical characteristics and rotational viscosity in the proximity of the bounding surface. Thus, we hope that our theoretical route will be useful for estimating the absolute values of these coefficients.

The classical approach for the viscosity of NLCs, in the framework of the EL theory, assumes that there is a unit vector field $\mathbf{n}(\mathbf{r}, t)$, representing the average alignment at each point of the field. The dynamic stress tensor (ST) $\bar{\sigma}$ of an incompressible NLC with velocity \mathbf{v} ($\nabla \cdot \mathbf{v} = 0$) is given by symmetric and antisymmetric parts [6]. Two relevant equations in our problem are (i) the balance of momentum and (ii) the vector equation for the director \mathbf{n} [1],

$$\rho \frac{d\mathbf{v}}{dt} = \nabla \cdot \bar{\sigma}, \quad (4)$$

$$\mathbf{T}_{vis} + \mathbf{T}_{el} + \mathbf{T}_{elast} = \mathbf{0}. \quad (5)$$

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where $\rho = N/V$ is the particle number density, the torque on the director due to viscous forces, $\mathbf{T}_{vis} = -\mathbf{n} \times (\gamma_1 \mathbf{N} - \gamma_2 \mathbf{M} \cdot \mathbf{n})$, the torque due to electrical forces $\mathbf{T}_{el} = (\epsilon_a/\epsilon_0) \mathbf{n} \times \mathbf{E}(\mathbf{E} \cdot \mathbf{n})$, and the torque due to elastic forces $\mathbf{T}_{elast} = \mathbf{n} \times \mathbf{h}$, with the molecular field $\mathbf{h} = \mathbf{h}_s + \mathbf{h}_t + \mathbf{h}_b$ being evaluated in terms of the Frank elastic coefficients $K_i (i=1,2,3)$ [6] for splay, twist, and bend deformations, respectively. Here $\mathbf{N} = d\mathbf{n}/dt - \mathbf{W} \cdot \mathbf{n}$, \mathbf{M} and \mathbf{W} are the symmetric and antisymmetric parts of the flow velocity gradient tensor $v_{i,j} = \partial v_i / \partial r_j$, and time derivative $d\mathbf{n}/dt = \partial \mathbf{n} / \partial t + \mathbf{v} \cdot \nabla \mathbf{n}$. The rotational viscosity coefficients are $\gamma_1 = \alpha_3 - \alpha_2$ and $\gamma_2 = \alpha_6 - \alpha_5$, where α_i are the Leslie coefficients. The components of the molecular field are given by [6] $\mathbf{h}_s = K_1 \nabla (\nabla \cdot \mathbf{n})$, $\mathbf{h}_t = -K_2 [(\mathbf{n} \cdot \mathbf{l}) \mathbf{l} + \nabla \times (\mathbf{n} \cdot \mathbf{l}) \mathbf{n}]$, $\mathbf{h}_b = K_3 [\mathbf{n} \times \mathbf{l} \times \mathbf{l} + \nabla \times (\mathbf{n} \times (\mathbf{n} \cdot \mathbf{l}))]$, where $\mathbf{l} = \nabla \times \mathbf{n}$, and $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ is the dielectric anisotropy of the NLC.

In the case of Couette flow the flow velocity $\mathbf{v} = (v_x(y), 0, 0)$ is in x direction, $\dot{\gamma} = \partial v_x / \partial y$ is the only non-vanishing component of the flow velocity gradient tensors M and W , and the director is aligned at the angle θ_{eff} , with components $n_x = \cos \theta_{eff}$ and $n_y = \sin \theta_{eff}$. The present study considers an incompressible fluid, which implies that the shear stress in the gradient direction and the shear rate are uniform in a planar shear flow. One can rewrite Eq. (5) in a dimensionless form as

$$\frac{\partial \theta_{eff}}{\partial \tau} + \frac{1}{2} \left[1 + \frac{\gamma_2}{\gamma_1} \cos 2\theta_{eff} \right] - \frac{\bar{B}^2}{2} \sin 2\theta_{eff} + h(\theta_{eff}) \frac{\partial^2 \theta_{eff}}{\partial \bar{y}^2} + \frac{1}{2} h'(\theta_{eff}) \left(\frac{\partial \theta_{eff}}{\partial \bar{y}} \right)^2 = 0, \quad (6)$$

where $\bar{B}^2 = B^2 / (\gamma_1 \dot{\gamma}) = [\epsilon_a \sigma^2 / (\bar{\epsilon}^2 \epsilon_0^3 \gamma_1 \dot{\gamma})] \exp(-2\bar{y})$, dimensionless y coordinate and time are $\bar{y} = y/\lambda_D$, and $\tau = \dot{\gamma} t$, respectively, $h(\theta_{eff}) = (K_1 \cos^2 \theta_{eff} + K_3 \sin^2 \theta_{eff}) / (\gamma_1 \dot{\gamma} \lambda_D^2)$, $h'(\theta_{eff})$ is the derivative of the $h(\theta_{eff})$ with respect to θ_{eff} . At high shear rates for 4-*n*-octyloxy-4'-cyanobiphenyl (8OCB) molecules and the surface charge density $\sigma = 10^{-3}$ C/m², the director orientation in the bulk is dominated by flow alignment and the influence of electric torque is restricted to boundary layers $0 \leq y \leq 3$ μm [see Eq. (3)]. At $\dot{\gamma} = 800$ s⁻¹, $|\mathbf{T}_{elast}| \sim 0.5$ N/m², $|\mathbf{T}_{vis}| \sim 4.0$ N/m², $|\mathbf{T}_{el}(y = 0.1 \mu\text{m})| \sim 212$ N/m², and $|\mathbf{T}_{el}(y = 1.0 \mu\text{m})| \sim 0.2$ N/m². Since the ratio $\Delta = |\mathbf{T}_{elast}| / |\mathbf{T}_{vis}|$ is 0.125, the elastic torque can safely be disregarded at shear rates $\dot{\gamma} \geq 800$ s⁻¹. At lower shear rates, for example at $\dot{\gamma} = 100$ s⁻¹, the values of $\Delta \sim 1$, and the director orientation is governed by the viscous, elastic, and electric forces, both in the bulk and close to the surface. In our calculations, $\lambda_s = 0.1$ μm is used in agreement with the experimental results for cyanobiphenyls at indium tin oxide (ITO) glass plate coated with organic materials [7].

For a stationary Couette flow ($\partial \theta_{eff} / \partial \tau = 0$) at arbitrary shear rates, Eq. (6) can be written as

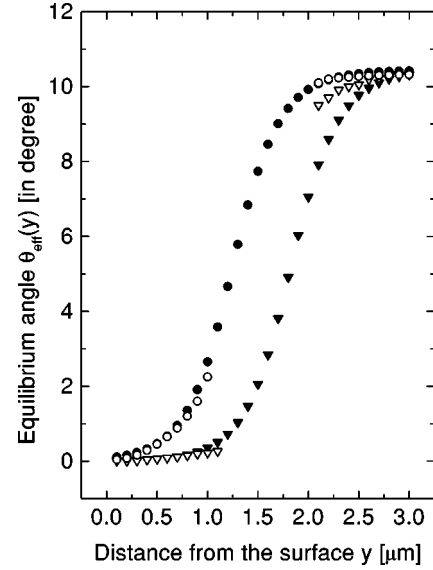


FIG. 1. The distance dependence of the flow alignment angle $\theta_{eff}(y)$, for homeotropic alignment ($\theta_s = 0$) of 8OCB molecules, at temperature $T = 340$ K, calculated using Eq. (11), at two values of the shear rate $\dot{\gamma}$; 100 s⁻¹ (solid triangles); 800 s⁻¹ (solid circles), and using Eqs. (8) and (9), at $\dot{\gamma} = 100$ s⁻¹ (open triangles); 800 s⁻¹ (open circles). The surface charge density is $\sigma = 10^{-3}$ C/m². The dimensionless interval for distance \bar{y} is $0.18 \leq \bar{y} \leq 5.5$.

$$\frac{\partial^2 \theta_{eff}(\bar{y})}{\partial \bar{y}^2} - \mathcal{A} \exp(-2\bar{y}) \theta_{eff}(\bar{y}) + \mathcal{B} = 0, \quad (7)$$

where the last term in Eq. (6) is negligible due to slowly varying $\theta_{eff}(\bar{y})$, $\mathcal{A} = (\epsilon_a / \bar{\epsilon}^2) (\sigma^2 / K_1) \lambda_D^2$, and $\mathcal{B} = [(\gamma_1 + \gamma_2) / 2] \dot{\gamma} \lambda_D^2 / K_1$. The boundary conditions are $\theta_{eff}(\bar{y}) = 0$ and $\partial \theta_{eff}(\bar{y}) / \partial \bar{y} = 0$, at $\bar{y} = \bar{y}_a = \lambda_s / \lambda_D$, and $\theta_{eff}(\bar{y}) = \theta_{bulk}$, and $\partial \theta_{eff}(\bar{y}) / \partial \bar{y} = 0$, at $\bar{y} = \bar{y}_b$. In our case $\bar{y}_b = 5.5$ (at $T = 340$ K). The justification of the choice in $\bar{y}_b = 5.5$ may be given by the distance dependence of the $\theta_{eff}(\bar{y})$, (see Fig. 1) which increases monotonically and saturates at $\bar{y}_b = 5.5$ ($y_b \sim 3$ μm). Near the wall the \mathbf{T}_{el} term dominates [which results in $\theta_{eff}(\bar{y}) = 0$ at $\bar{y} = \bar{y}_a$, both for homeotropic ($\theta_s = 0$) and planar alignments ($\theta_s = \pi/2$)]; on the other hand, at $\dot{\gamma} \leq 100$ s⁻¹, both \mathbf{T}_{elast} and \mathbf{T}_{vis} terms dominate far from the wall. Thus, Eq. (7) is solved in three separate regions; first, close to the \bar{y}_a , second, close to the \bar{y}_b , and third, in the interval $\bar{y}_a \leq \bar{y} \leq \bar{y}_b$. Near $\bar{y} \sim \bar{y}_a = \lambda_s / \lambda_D = 0.18$, one may determine a power series solution of Eq. (7) in the form

$$\theta_{eff}(\bar{y}) = \frac{\mathcal{B}}{\mathcal{A} \bar{y}_a^2 - 2} (\bar{y}_a - \bar{y})^2. \quad (8)$$

A similar solution is obtained near $\bar{y} \sim \bar{y}_b$, where $|\mathcal{A} \exp(-2\bar{y})| \sim 0.006$ can be discarded, one may determine a solution of Eq. (7) in the form

$$\theta_{eff}(\bar{y}) = \theta_{bulk} + \frac{\mathcal{B}}{2}(\bar{y}_b - \bar{y})^2. \quad (9)$$

It should be noted that both these solutions, Eqs. (8) and (9), satisfy the above mentioned boundary conditions. Solution in the intermediate range ($\bar{y}_a \ll \bar{y} \ll \bar{y}_b$) may be obtained by asymptotically matching these two solutions.

For a stationary Couette flow with high shear rates $\dot{\gamma} \geq 800 \text{ s}^{-1}$, Eq. (6) leads to [1]

$$\dot{\gamma}[\gamma_1 + \gamma_2 \cos 2\theta_{eff}(\bar{y})] = B^2 \sin 2\theta_{eff}(\bar{y}). \quad (10)$$

Thus, the torque balance yields a flow alignment angle

$$\cos 2\theta_{eff}(\bar{y}) = \lambda_{eff}^{-1} = \alpha_0 \kappa [1 + \{1 + (C^4 - 1)\kappa^{-1}\}^{1/2}], \quad (11)$$

where $\alpha_0 = \lambda^{-1} = -\gamma_1/\gamma_2$, $\kappa = (1 + \alpha_0^2 C^4)^{-1}$, $C^2 = B^2/\gamma_1 \dot{\gamma}$. Taking into account that the RVCs γ_i ($i=1,2$) can be expressed according to Kuzuu and Doi (KD) theory [11] as

$$\gamma_i = \begin{cases} \frac{2f}{\lambda} \bar{P}_2, & i=1, \\ -2f, & i=2, \end{cases}$$

where \bar{P}_2 is the second-rank nematic order parameter (OP), $f = (\rho k_B T / D_\perp) p$, D_\perp is the rotational diffusion coefficient for molecular tumbling motions, k_B is the Boltzmann constant, T is the temperature, p is a particle geometric factor usually taken as $p = (a^2 - 1)/(a^2 + 1)$, a is the length-to-breadth ratio of the molecule, the ratio γ_1^{eff}/γ_1 is obtained from Eq. (11) as

$$\frac{\gamma_1^{eff}}{\gamma_1} = \frac{\lambda}{\lambda_{eff}} = \kappa [1 + \{1 + (C^4 - 1)\kappa^{-1}\}^{1/2}]. \quad (12)$$

Here γ_1^{eff} is the effective RVC in the presence of the charged surface. We note that the bounding surface influences \bar{P}_2 only in a thin bulk layer ($\sim 10\text{--}100 \text{ nm}$) [2,7] close to the surface and \bar{P}_2 is not affected by the flow, except for temperatures very close to the isotropic-nematic phase transition [12]. Recently, the role of surface coupling on D_\perp at the LC-solid interface has been investigated by means of broadband dielectric spectroscopy [13]. It was found that the frequencies for the hindered rotation of molecules around their molecular short axes remain unchanged in cylindrical channels of Anopore membranes having a diameter up to $0.2 \mu\text{m}$ with respect to the bulk phase, at least for *cyanobiphenyls*. Since the relaxation time of the hindered rotation $\tau_{00}^1 = [D_\perp(2 - 2\bar{P}_2)/(1 - 2\bar{P}_2)]^{-1}$ [14], one can take D_\perp and \bar{P}_2 at $y > \lambda_s$ being only temperature dependent. According to the KD theory the charged surface, over the distance more than λ_s , can influence only two of the six Leslie coefficients α_2 and α_3 (and γ_1) due to its influence on the asymmetric part of the ST.

In order to examine the effect of the charged surface on the flow alignment angle $\theta_{eff}(\bar{y})$ and γ_1^{eff} , we consider the

nematic phase of 8OCB on an ITO-coated glass plate. Recently, the D_\perp , the bulk RVCs γ_i ($i=1,2$), and the OPs \bar{P}_{2L} ($L=1,2,3$) have been calculated for 8OCB [15]. It was done by combining the existing statistical-mechanical approach [16,17] and nuclear magnetic resonance relaxation theory [18]. The values of $\bar{\epsilon}$ and ϵ_a are determined using the temperature dependent coefficients ϵ_\parallel and ϵ_\perp for 8OCB obtained in Ref. [19]. Taking into account that the bulk ion concentration $n_{bulk} = N_+/V = N_-/V \approx 5 \times 10^{20} \text{ m}^{-3}$ [20,21], where N_+ and N_- represent the number of positive and negative ions, respectively [for a weak electrolyte ($N_+ \approx N_-$)], one can calculate the bulk Debye screening length $\lambda_D = (\epsilon_0 \epsilon_\parallel k_B T / 2e^2 n_{bulk})^{1/2}$ [20], where $e = 1.602 \times 10^{-19} \text{ C}$ is the proton charge, and ϵ_\parallel is the dielectric constant parallel to the director \mathbf{n} . In the temperature range where the nematic phase of 8OCB exists, one has $\lambda_D \in [0.545, 0.556] (\mu\text{m})$ for homeotropic alignment. In our calculations the range of shear rate is $\dot{\gamma} \in [100, 800] (\text{s}^{-1})$ [22,23]. Figure 1 shows the distance dependence of the $\theta_{eff}(y)$, calculated due to Eqs. (8), (9), and (11), for homeotropically aligned 8OCB molecules with $\sigma = 10^{-3} \text{ C/m}^2$ at $T = 340 \text{ K}$. At this temperature, the equilibrium angle $\theta_{eff}(y)$, increases monotonically with distance y up to the bulk value of $\theta_{eff}(y) = \theta_{bulk} = 10.35^\circ$. With an increasing shear rate $\dot{\gamma}$ from 100 s^{-1} up to 800 s^{-1} , deviations between the flow alignment angle $\theta_{eff}(\bar{y})$, calculated from Eqs. (9) and (11), at the right end of the interval $[\bar{y}_a, \bar{y}_b]$ decrease as expected, whereas at the left end of the same interval such deviation is negligible, both for high and lower values of the shear rates. This shows that the influence of the elastic torque on the spatial dependence of $\theta_{eff}(\bar{y})$ is vanishingly small up to $y \sim 1 \mu\text{m}$, and both Eqs. (8) and (11) give, practically, the same results. At larger distances $1 < y < 2.75 \mu\text{m}$, the director orientation is governed by the electric, viscous, and elastic forces for $\dot{\gamma} \sim 100 \text{ s}^{-1}$, and Eq. (11) is, therefore, inappropriate. Only for $\dot{\gamma} \geq 800 \text{ s}^{-1}$, the elastic forces can safely be neglected. The distance dependent γ_1^{eff}/γ_1 for the homeotropic alignment of 8OCB molecules at an ITO-coated glass plate and at a fixed surface charge density 10^{-3} C/m^2 is shown for two temperatures ($T = 340, 350 \text{ K}$) in Fig. 2. It is clear that the surface electric field has a larger effect on γ_1^{eff} as the temperature decreases. With decreasing distance y , the effect of the charged surface on γ_1^{eff} increases monotonically, and its effect vanishes at distance $y \sim 3 \mu\text{m}$. The presence of a charged surface gives an additional contribution to the γ_1^{eff} up to 7.8% (340 K). Recently, the absolute values of the RVCs γ_i ($i=1,2$), in the bulk nematic phase of 8OCB has been calculated [15]. The γ_1 values are 4.68×10^{-2} , and $1.72 \times 10^{-2} \text{ Pa s}$ at 340, and 350 K, respectively. Thus the effective RVC γ_1^{eff} can be calculated. It should be noted that according to Eq. (12) the surface charge density $\sigma \rightarrow \infty$ leads to $\lim_{\sigma \rightarrow \infty} \gamma_1^{eff}/\gamma_1 = \alpha_0^{-1} = -\gamma_2/\gamma_1$, with simultaneous growth of the penetration length over which the charged surface can disturb the bulk nematic. The largest contribution to the rotational viscosity, due to the charged surface, is about 6–18%, at least for *cyanobiphenyls* [24], except at tempera-

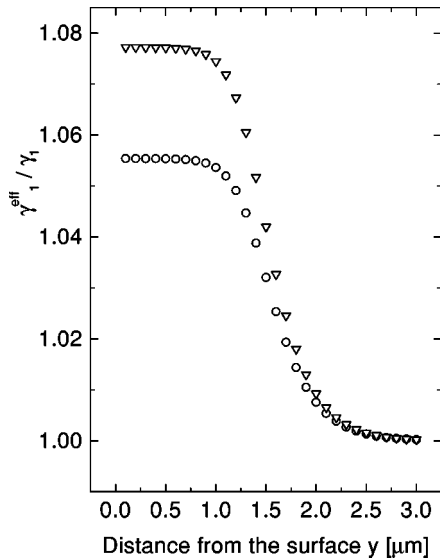


FIG. 2. The distance dependence of the ratio γ_1^{eff}/γ_1 , for homeotropic alignment of 8OCB molecules, at the surface charge density $\sigma=10^{-3}$ C/m² and $\dot{\gamma}=800$ s⁻¹, calculated using Eq. (12), at two different temperatures: $T=340$ K (triangles), and 350 K (circles).

tures close to the nematic-smectic transition [25]. When the surface charge density $\sigma \rightarrow 0$, $\lim_{\sigma \rightarrow 0} \gamma_1^{eff}/\gamma_1 = 1$.

In summary, we have investigated the influence of the charged bounding surface on the flow alignment angle (at shear rates $100 \leq \dot{\gamma}/s^{-1} \leq 800$), and rotational viscosity of a liquid crystal, 4-*n*-octyloxy-4'-cyanobiphenyl, using the conventional EL theory. The role of charged surfaces is ac-

counted only by the long-range component (due to the surface electric field) of the surface potential. The effective enhancement of rotational viscosity in shear flowing LCs near the charged bounding surface is interpreted in terms of the orientational change of the director caused by the surface electric field. A balance between the frictional and electric torques exerted on the director is reflected in a growth of the effective rotational viscosity up to 7.8% (at 340 K) with respect to the bulk value. The temperature dependence of γ_1^{eff} also supports this conclusion. We note that only at a shear rate $\dot{\gamma} \sim 100$ [s⁻¹], the director orientation is governed both by the elastic ($|\mathbf{T}_{elast}| \sim 0.5$ N/m²) and viscous ($|\mathbf{T}_{vis}| \sim 0.5$ N/m²) torques. Our analysis also shows that the influence of both the elastic and viscous forces are vanishingly small up to 0.8 μm for the studied surface field. For y larger than 0.8 μm , the ratio γ_1^{eff}/γ_1 should be calculated using Eq. (7) for shear rates $\dot{\gamma} \sim 100$ s⁻¹. It should be noted that the present results for flow aligning nematics composed of prolate particles with positive dielectric anisotropy are valid, according to Eq. (12), also for systems composed of both prolate or oblate molecules with negative dielectric anisotropy. The flow alignment angle is influenced by the large axial dipole moment in a cyanobiphenyl molecule, which produces high values of dielectric constants [26] in the factor C and Debye screening length λ_D . We believe that the present study not only shows some useful routes for estimating the viscosity of cyanobiphenyls and for analyzing the electrorheology of NLCs, but also clarifies the role of elastic torques in flow alignments near charged surfaces.

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