PHYSICAL REVIEW E **89**, 022509 (2014)

Lehmann rotation of the cholesteric helix in droplets oriented by an electric field

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(Received 7 January 2014; published 21 February 2014)

We study the Lehmann rotation of the cholesteric helix in droplets of the liquid crystal *N*-(*p*-methoxybenzilidene)-*p*-butylaniline doped with a small amount of the chiral molecule R811 when they are subjected to a temperature gradient. We show that the helix rotates much faster when it is parallel to the temperature gradient than when it is perpendicular to it. The first configuration is obtained by submitting the droplets to an ac electric field parallel to the temperature gradient, whereas the second one is observed at zero field. We show that the rotation velocity of the helix strongly depends on the droplet radius, even when the helix is parallel to the temperature gradient. This observation supports the idea that the Leslie thermomechanical coupling cannot explain alone the Lehmann effect.

DOI: 10.1103/PhysRevE.89.022509 PACS number(s): 61.30.−v, 65.40.De, 05.70.Ln

I. INTRODUCTION

In 1900, Lehmann [1] observed the continuous rotation of the internal texture of cholesteric droplets when subjected to a temperature gradient. The experiment was first reproduced in 2008 by using a compensated cholesteric (mixture of octyloxy-cyanobiphenyl and cholesteryl chloride in equal proportions) [2,3] and 1 year later by using diluted cholesteric mixtures (i.e., nematic phase doped with a small amount of chiral molecules) [4,5]. In all of these experiments, the droplets coexist with their isotropic liquid and have a banded texture, indicating that the helical axis is rather perpendicular to the temperature gradient. The rotation of the texture was explained within the model of Leslie taking into account the chiral terms in the constitutive equations of the cholesteric phase [6]. According to this model, the director experiences a torque proportional to the temperature gradient $\vec{G}$ of the expression

$$\vec{\Gamma}_{\text{Leslie}} = \nu\vec{n}\times(\vec{n}\times\vec{G}), \qquad (1)$$

where ν is the so-called Lehmann coefficient. By writing that this torque equilibrates with the viscous torque and by assuming that there is no flow of matter (we will come back later to this assumption), it was shown that the droplets rotate with an angular velocity ω given by [2,3]

$$-\frac{\nu G}{\gamma_1\omega} = 1 + \frac{\iiint_{\text{drop}}\left[\vec{e}_z\cdot\frac{\partial\vec{n}}{\partial\theta}\times\vec{n} + \left(\frac{\partial\vec{n}}{\partial\theta}\right)^2\right]dV}{\iiint_{\text{drop}}\left[\vec{e}_z\cdot\frac{\partial\vec{n}}{\partial\theta}\times\vec{n} + 1 - (\vec{e}_z\cdot\vec{n})^2\right]dV}. \qquad (2)$$

In this expression, $\vec{e}_z$ is the unit vector parallel to the temperature gradient, θ the polar angle, and γ_1 the rotational viscosity. This equation shows that the texture angular velocity is proportional to G and has the form $\omega = -(1+A)\nu G/\gamma_1$, where A is a dimensionless coefficient which depends on the director field and on the drop geometry. This coefficient was calculated by making different assumptions on the shape of the droplet (flattened cylindrical box or spherical cap [7,8]) and the structure of the director field (perfect helix [7,8] or helix strongly deformed by the the anchoring conditions at the surface of the droplet [2]). In all cases, it was shown that A was an even and rapidly increasing function of the product kR, where k is the wave vector of the banded texture (in practice, always close to q, the equilibrium twist of the phase) and R the radius of the droplet. In addition, it was shown that $A \to 0$ when $R \to 0$. As a consequence, it is sufficient to extrapolate the curve $\omega(R)/G$ to $R = 0$ to obtain the ratio ν/γ_1. The application of this procedure to different cholesteric mixtures showed that the Lehmann coefficient ν is proportional to the equilibrium twist (and so to the concentration of chiral molecules in diluted cholesteric mixtures).

On the other hand, a very surprising result was that the coefficient ν obtained in this way was systematically much larger than the coefficient ν measured below the transition temperature in the Leslie geometry (i.e., when the helix is parallel to the temperature gradient and can freely rotate on the glass plates limiting the sample) [7–9]. Our conclusion was that the coefficient ν obtained from the droplet rotation was perhaps not the true Leslie coefficient given by Eq. (1), but an effective coefficient taking into account other phenomena neglected in the calculation leading to Eq. (2), such as a possible macroscopic flow.

In this paper, we propose to test again the simplified model of Refs. [2,3] by changing the orientation of the helix in the droplets. For this purpose, we use a cholesteric liquid crystal of negative dielectric anisotropy and apply an ac electric field parallel to the temperature gradient to change the orientation of the helix. In this way, the helix can be oriented parallel to the temperature gradient inside the droplets providing that the electric field is large enough. In that case, the director orientation must become independent of the polar angle θ if the director is not strongly anchored on the contour of the droplet and $A = 0$. Thus, the simplified model predicts that the helix should rotate at constant speed $\omega = -\nu G/\gamma_1$ inside the droplets, whatever their radii. We propose to test this prediction experimentally and to compare the rotation velocity of the helix depending on whether it is parallel or perpendicular to the temperature gradient.

II. EXPERIMENTAL DETAILS

The liquid crystal chosen was *N*-(*p*-methoxybenzilidene)-*p*-butylaniline (MBBA from Frinton Laboratories, Inc)

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and the chiral dopant was R-(+)-octan-2-yl 4-((4-(hexyloxy)benzoyl)oxy) benzoate (R811 from Merck Ltd). We prepared three mixtures with concentration C of R811 of 0.5, 1, and 2 wt%, respectively. The helical twist power of R811 [HTP defined to be $1/(PC)$ with P the pitch and C the concentration in wt%] was measured at the transition temperature to the isotropic liquid by using the Cano-wedge method. We found HTP $= 9.6\ \mu\text{m}^{-1}\ \text{wt\%}^{-1}$. The samples, of thickness 30 μm, were prepared between two parallel glass plates coated with ITO (tin-doped indium oxide) and a thin layer of a polymercaptan, a surface treatment that gives a planar and sliding anchoring (for more details see Refs. [10,11]). The setup used to impose a temperature gradient perpendicular to the glass plates was described in a previous publication [2]. In the following, we denote by ΔT the difference of temperature between the top and bottom ovens. To a good approximation, the temperature gradient is given by $G = \frac{\Delta T}{4e}\frac{\kappa_g}{\kappa_{\text{LC}}}$, where e is the thickness of the glass plates (1 mm) and $\kappa_g/\kappa_{\text{LC}}$ the ratio of the conductivities of the glass and of the liquid crystal, close to 3 by taking $\kappa_g \approx 1.1\ \text{Wm}^{-1}\ \text{s}^{-1}$ [12] and $\kappa_{\text{LC}} \approx 0.37\ \text{Wm}^{-1}\ \text{s}^{-1}$ [13,14]. With these values, one obtains $G\ (\text{Km}^{-1}) \approx 0.012\Delta T$ (K). A function generator HP3325B coupled to a homemade amplifier (×20; response time, 0.5 μs) is used to impose the large amplitude ac electric field necessary to change the orientation of the helix.

III. DROPLET ROTATION WITHOUT ELECTRIC FIELD

We first performed the experiment without an electric field. A few droplets are shown in Fig. 1. They are quite similar to the droplets observed in our previous experiments with biphenyls. Their banded structure of wavelength close to $P/2$ indicates that the helix is little deformed and aligns perpendicularly to the temperature gradient. In this figure, the droplets rotate clockwise while the temperature gradient points towards the observer which means that their rotation vector and the temperature gradient are oriented in opposite directions. As a consequence, $\nu > 0$, which was expected for R811 [4,5]. For each concentration, we measured the rotation period $\Theta_\perp = 2\pi/\omega_\perp$ of the droplets as a function of their radius R and the temperature difference ΔT. The subscript $\perp$ indicates that in these experiments, the helix is perpendicular to the temperature gradient. As can be seen in Fig. 2(a), the data obtained at each concentration collapse on a single curve on the condition to plot the product $\Theta_\perp \Delta T$ as a function of R. This shows that $\omega \propto G$ at fixed radius, in agreement with the simplified model [see Eq. (1)]. On the other hand, it was not possible to collapse all the data onto a universal curve (in particular the data obtained at the smallest concentration of 0.5 wt%) by plotting $\Theta_\perp C \Delta T$ as a function of qR [with $q = 2\pi\text{HTP} \times C$] [Fig. 2(b)], as the simplified model suggests. This indicates that the function $A(qR)$ changes with the concentration, in particular, when the drop thickness becomes comparable to or smaller than the equilibrium pitch, what happens at the lowest concentration. In this case, the helix partly unwinds and the simplified model predicts a lower dissipation than when the helix is not deformed. This could qualitatively explain why the droplets observed in the mixture at 0.5 wt% rotate comparatively faster than the droplets of similar radii observed in the two other mixtures at 1 and 2 wt%. Unfortunately, the data are too much dispersed to allow a confident extrapolation of the data at zero radius and a serious determination of the Lehmann coefficient ν. This dispersion, which is larger than in our experiments with the biphenyls, is mainly due to a pollution of the MBBA with the polymercaptan. This pollution leads to a continuous decrease of the melting temperature during the experiments, in particular, during the first hours following the filling of the samples. This effect is accompanied by a viscosity increase [15] that leads to a slowing down of the droplets (of the order of

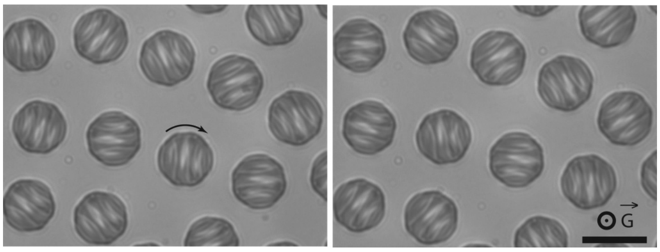

FIG. 1. Droplets of the cholesteric phase which coexist with the isotropic liquid in the absence of an electric field. The time interval between the two pictures taken in natural light is equal to 21 s. $C = 1$ wt% and $\Delta T = 10\,^\circ$C. The arrow indicates the sense of rotation. The bar is 30 μm long.

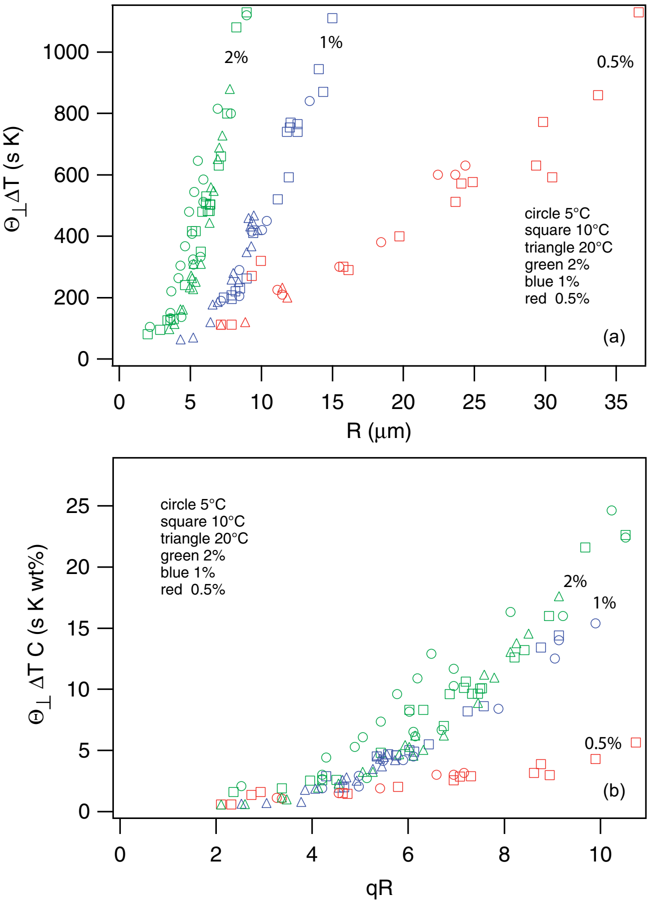

FIG. 2. (Color online) (a) Product $\Theta_\perp \Delta T$ as a function of R. (b) Product $\Theta_\perp \Delta T C$ as a function of qR.

20% for a temperature decrease of 3–4 °C). Experimentally, we stop measurements with the same sample when its transition temperature has decreased by more than 5 °C, which happens typically 3 days after filling.

IV. DROPLET ROTATION UNDER ELECTRIC FIELD

In order to orient the helix parallel to the temperature gradient, we submitted the droplets to a large ac electric field (we recall that MBBA is of negative dielectric anisotropy [16]). In practice, a voltage of amplitude 40 Vrms was largely enough to produce this effect in all samples. In order to avoid convective instabilities, we worked at high frequency, typically 10 kHz. We checked that all the results presented in this section were completely independent of the frequency and the amplitude of the voltage (which we changed from 2 to 100 kHz and from 30 to 150 Vrms, respectively). This indicates that the only effect of the field is to orient the helix parallel to the temperature gradient. When the field is applied, the banded texture disappears and no rotation can be detected in natural light. In order to detect a helix rotation, the sample must be placed between crossed polarizers. Doing this, we first observed that the transmitted intensity was only a function of the distance r from the center of the droplets. This indicates that the helix orientation is independent of the polar angle θ. On the other hand, we cannot say whether the intensity variation along r is only due to the droplet thickness variation along r or is also due to a disorientation of the helix as a function of r, as these two phenomena produce the same effect. Then, we observed that the intensity oscillated inside the droplets as can be seen in Fig. 3. This clearly indicates that the helix rotates with a period equal to 4 times the period of the optical signal (this can be easily checked from the complete calculation of the transmitted intensity given in Ref. [17], p. 126). By rotating the polarizers, we checked that the rotation vector of the helix and the temperature gradient pointed in opposite directions, which indicates that $\nu > 0$. We then systematically measured the period of rotation of the helix $\Theta_\parallel$ as a function of R, ΔT, and C. As before, we found that it was possible to collapse all the data measured at a given concentration by plotting $\Theta_\parallel \Delta T$ as a function of R [Fig. 4(a)]. In this case, the dependence on R is linear. More interestingly, and in contrast with the experiments without an electric field, we found that it was possible to collapse all the data onto a single line by plotting $\Theta_\parallel \Delta T C$ as a function of R [Fig. 4(b)]. This result is particularly interesting in many respects. First, it shows that, as expected, the helix rotation velocity measured in droplets is proportional to the temperature gradient and to the concentration of chiral molecules. Second, it shows that the helix rotation velocity is inversely proportional to the droplet radius. This dependence, on the other hand, is surprising because it disagrees with the simplified model of Ref. [2] that predicts a constant rotation velocity: $\omega_\parallel = \nu G/\gamma_1$ according to Eq. (1). We nevertheless note that the helix rotation velocity extrapolated at $R = 0$ from these measurements is compatible with the rotation velocity of the smallest droplets observed in zero field. This is normal because the orientation of the director must be the same in these two limiting cases.

Finally, we performed an experiment in the Leslie geometry [6] by preparing a planar sample just below the melting temperature. In that case, the cholesteric layer is in contact with the two polymercaptan layers and the electric field

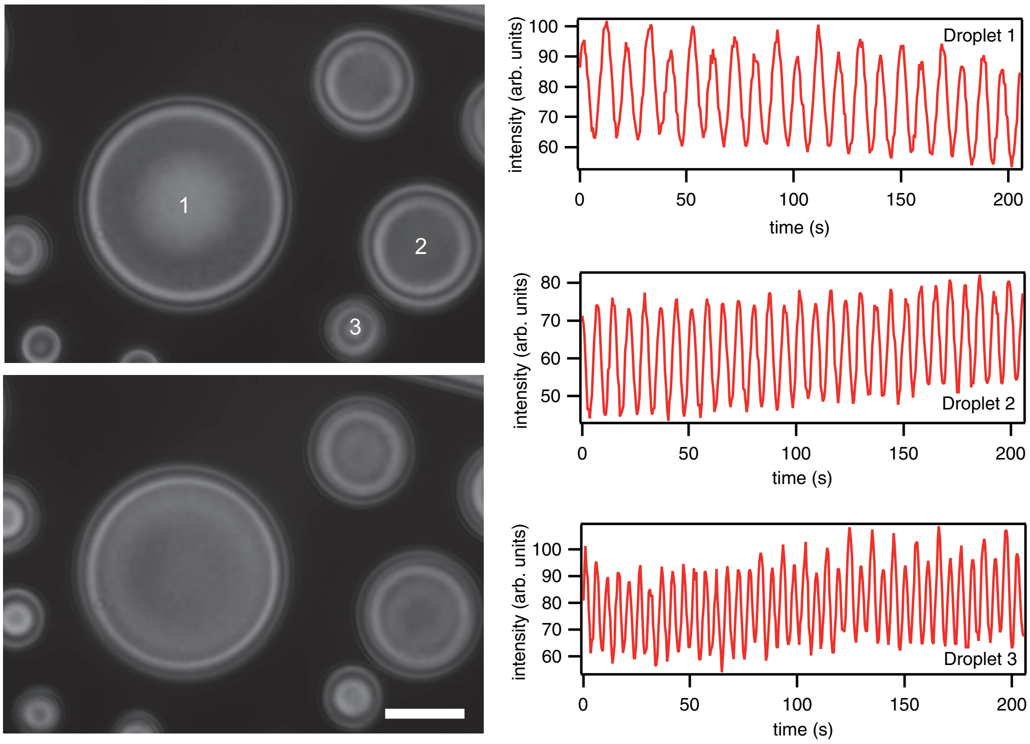

FIG. 3. (Color online) Droplets observed between crossed polarizers under the electric field (10 kHz, 40 Vrms). The top photograph was taken at time $t = 12.5$ s and the bottom one at time $t = 17.5$ s. The three graphs show the intensity measured in the middle of the droplets marked 1, 2, and 3 in the top photograph. The bar is 30 μm long.

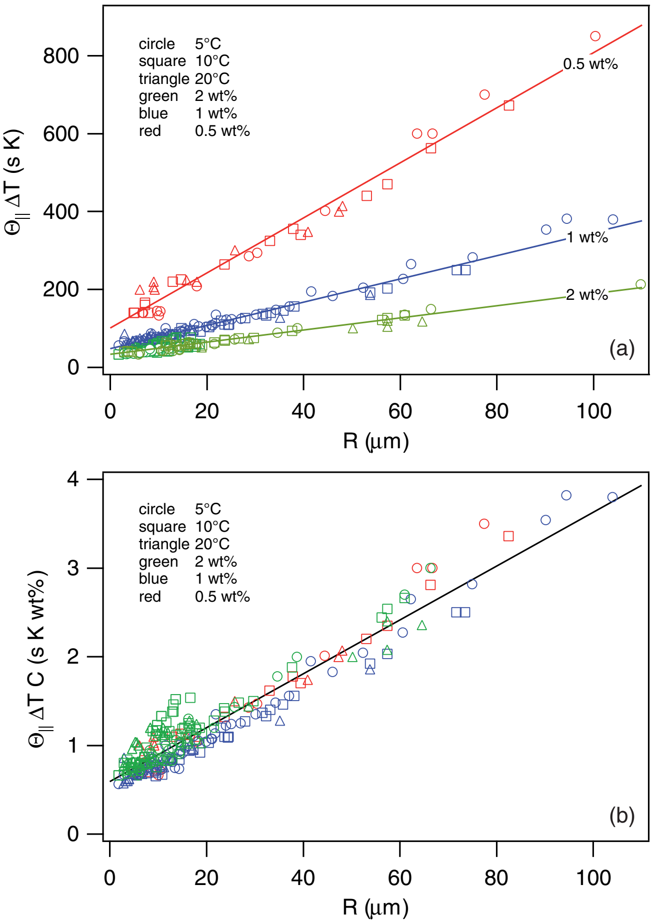

FIG. 4. (Color online) (a) Product $\Theta_{\parallel}\Delta T$ as a function of R. (b) Product $\Theta_{\parallel}\Delta T C$ as a function of R.

may be suppressed while keeping the helix parallel to the temperature gradient. We observed that in all samples, the helix was not visibly rotating. This indicates that the Lehmann coefficient ν (one should rather speak of the thermomechanical Leslie coefficient ν as we know that we really measure this coefficient in this geometry) is extremely small and almost impossible to measure below the transition temperature. This result is compatible with our previous measurements in the diluted mixtures made with biphenyl liquid crystals [8]. The same behavior was observed by partly melting the cholesteric layer from the hot plate. In that case, the planar orientation was maintained by applying an electric field (if not the structure destabilizes and bands form). This result is obviously compatible with the fact that in droplets, $\omega_{\parallel} \propto 1/R$ and thus tends to 0 when R diverges.

V. FEEDBACK ON THE THEORETICAL MODEL

These new results clearly show that the simplified model of Ref. [2] is, if not wrong, at least incomplete. One reason could be that this model implicitly supposes that there is no flow. This is the case in the Leslie geometry when the cholesteric layer has an infinite extension in the horizontal plane and the helix is parallel to the temperature gradient [6]. On the other hand, there are certainly flows in the droplets when the helix is perpendicular to the temperature gradient, but the calculations are impossible to carry out analytically in this geometry. The same question obviously arises in the droplets under an electric field when the helix is parallel to the temperature gradient. In particular, one could ask whether the boundary conditions on the side surfaces of the droplets are satisfied in the simplified model and, if not, if they are responsible for the variation of the helix rotation velocity observed in this geometry when the droplet radius changes.

To answer this question, we assume that the droplets have the shape of cylindrical boxes of radius R in which the helix is oriented parallel to the temperature gradient and we assume that the Leslie solution is still valid inside. Let ϕ be the angle between the director $\vec{n}$ and the x axis. According to Leslie [6], ϕ only depends on z and t and reads

$$\phi = qz + \omega t. \tag{3}$$

Substitution into the bulk torque equation

$$\gamma_1 \frac{\partial \phi}{\partial t} = K_2 \frac{\partial^2 \phi}{\partial z^2} - \nu G \tag{4}$$

gives the angular rotation velocity: $\omega = -\frac{\nu G}{\gamma_1}$. If the anchoring is fully sliding on the surface of the droplet (no anchoring and no surface viscosity, which are the assumptions made in the model of Ref. [2]), this solution also satisfies the surface torque equation on the surface of the droplet. This solution must also satisfy the bulk linear momentum equation and the associated boundary conditions which read

$$\text{div}\underline{\sigma} = 0 \tag{5}$$

in the bulk and

$$\underline{\sigma}\vec{\kappa} = -P_I\vec{\kappa} \tag{6}$$

on the surface (in the absence of flows), by denoting by $\vec{\kappa}$ the unit vector normal to the surface and pointing outwards and by P_I the pressure in the isotropic liquid. Because the helix is not distorted, the elastic stress tensor $\underline{\sigma}^e$ vanishes so that the total stress tensor reduces to

$$\underline{\sigma} = -P\underline{I} + \underline{\sigma}^v. \tag{7}$$

Here, P is the hydrostatic pressure and $\underline{\sigma}^v$ the viscous stress tensor of components in the absence of flow [18]:

$$\sigma^v_{ij} = \alpha_2 n_j \dot{n}_i + \alpha_3 n_i \dot{n}_j - \mu_1 (\vec{G}\cdot\vec{n})_i n_j - \mu_2 (\vec{G}\cdot\vec{n})_j n_i, \tag{8}$$

where $\vec{G} = G\vec{e}_z$ is the temperature gradient, α_2 and α_3 are two of the five Leslie viscosities (with $\gamma_1 = \alpha_3 - \alpha_2$), and μ_1 and μ_2 are the two thermomechanical Leslie coefficients (with $\nu = \mu_1 - \mu_2$).

From Eqs. (3) and (8), we calculate

$$\begin{aligned}
\sigma^v_{xx} &= -\sigma^v_{yy} = \tfrac{1}{2}\sin(2\phi)[G(\mu_1+\mu_2) - \omega(\alpha_2+\alpha_3)],\\
\sigma^v_{xy} &= (\mu_1 G - \alpha_2\omega)\sin^2\phi - (\mu_2 G - \alpha_3\omega)\cos^2\phi,\\
\sigma^v_{yx} &= (\mu_2 G - \alpha_3\omega)\sin^2\phi - (\mu_1 G - \alpha_2\omega)\cos^2\phi,\\
\sigma^v_{xz} &= \sigma^v_{zx} = \sigma^v_{yz} = \sigma^v_{zy} = \sigma^v_{zz} = 0.
\end{aligned} \tag{9}$$

As ϕ only depends on z, it comes $\text{div}\underline{\sigma}^v = 0$ so that $P =$ constant according to Eq. (5).

Finally, the application of Eq. (6) on the horizontal faces of the droplet gives $P = P_I$, while its application on the vertical side gives

$$\begin{aligned} \sigma^v_{xx}\kappa_x + \sigma^v_{xy}\kappa_y &= 0, \\ \sigma^v_{yx}\kappa_x + \sigma^v_{yy}\kappa_y &= 0. \end{aligned} \tag{10}$$

The stress field given in Eq. (9) does not satisfy these boundary conditions on the vertical side of the droplet at each point, but only on average along z. Indeed, averaging along z gives $\langle \sin(2\phi)\rangle = 0$ and $\langle \sin^2\phi\rangle = \langle \cos^2\phi\rangle = \frac{1}{2}$, so that $\langle \sigma^v_{xx}\rangle = \langle \sigma^v_{yy}\rangle = 0$ and $\langle \sigma^v_{xy}\rangle = \langle \sigma^v_{yx}\rangle = \frac{1}{2}(\nu G + \gamma_1\omega)$. The boundary conditions (10) are thus satisfied on average along z if $\omega = -\frac{\nu G}{\gamma_1}$. This velocity is the same as the one obtained from the bulk torque equation (4).

Another way to calculate ω consists of applying the angular momentum theorem to the droplet. This gives

$$\frac{d\vec{M}}{dt} = \int_S (\vec{r} \times \underline{\sigma}\vec{\kappa} + \underline{C}\vec{\kappa})dS, \tag{11}$$

where $\vec{M}$ is the angular momentum, $\underline{\sigma} = -P\underline{I} + \underline{\sigma}^e + \underline{\sigma}^v$ the total stress tensor, $\underline{C}\vec{\kappa}$ the surface elastic torque, and $\vec{\kappa}$ the unit vector pointing outwards of the droplet. Note that the right-hand side of this equation is nothing else than the torque that the isotropic liquid exerts on the surface of the droplet. In the stationary regime, this torque must vanish (in the absence of external forces) because $d\vec{M}/dt = 0$. This is compatible with our assumption that the isotropic liquid is at rest. In our model, we assume that the helix is not distorted. As a consequence, $\underline{C}$ and the elastic stress tensor $\underline{\sigma}^e$ also vanish. Finally, we must have

$$\int_S (\vec{r} \times \underline{\sigma}^v\vec{\kappa})dS = 0, \tag{12}$$

where the components of the viscous stress tensor $\underline{\sigma}^v$ are given in Eq. (9). A straightforward calculation shows that this equation is satisfied (whatever the height of the droplet along z) if $\omega = -\frac{\nu G}{\gamma_1}$.

Again, we find the same velocity as the one obtained from the bulk torque equation (4).

For this reason, we think that this approximate solution is close to the exact solution for a droplet of finite size. The problem, on the other hand, is that this solution predicts a constant rotation velocity of the helix whatever the radius of the droplet, which is in complete disagreement with the experiment (see Fig. 4).

The fact that the isotropic liquid exerts no torque on the surface of the droplet may seem surprising at first sight if one thinks about the apparently similar problem of a droplet subjected to a rotating magnetic field. In this case, the director also rotates under the action of the magnetic torque, but we know that the nematic phase exerts on its surface a torque equal to the torque that the magnetic field exerts on the bulk nematic phase [19,20]. This is due to the fact that the magnetic torque is an external torque [it must be added in the right-hand side of Eq. (11)], whereas the Leslie torque is an internal torque.

For the moment, we cannot say with certainty whether there is a flow in or out of the droplets. So far, we assumed that there is no flow and that the rotation of the texture was only due to the rotation of the molecules around their centers of gravity at angular velocity ω. Nonetheless, a rigid rotation of the droplets at angular velocity ω could explain as well our optical observations. If such a solution exists, the isotropic liquid should be sheared in the vicinity of the droplets, which means that each droplet should exert a torque different from 0 on the isotropic liquid. The previous calculation shows that this hypothesis is incompatible with the Leslie model.

As a consequence, the temperature gradient does not act, at least in the Leslie model, as a rotating magnetic field or a rotating electric field, even if the phenomena seem to be similar at first sight. We emphasize that the same conclusion holds in the case of a nematic droplet subjected to the electric field of a circularly polarized laser light. In this case, the droplets have been shown to rotate to a good approximation as rigid bodies [21]. This is not incompatible with our analysis because the droplets are here submitted to a torque different from 0 due the external field of the laser light.

VI. CONCLUSION

To conclude this paper, we would like to stress that the droplets rotate independently of each other, even when they are almost in contact, close to coalescence. In particular, two neighboring droplets never turn on a circle as two vortices with identical circulation would do in a fluid at rest at infinity. This observation and the fact that we never detected any motion of dust particles in the isotropic liquid close to the droplets suggest that there is no flow in the isotropic liquid, which would imply that the droplets do not rotate as rigid bodies. This remark is compatible with the Leslie theory and the observations of Lehmann who pointed out that it is the internal texture of the droplets which is rotating and not the droplets themselves. On the other hand, the Leslie thermomechanical coupling seems to be not strong enough to explain alone the observations. In addition, it fails to explain the decrease of the rotation speed of the helix in the droplets oriented under an electric field. For these two reasons, we suspect that another, more efficient, mechanism exists to explain the Lehmann effect. One possibility could be a Marangoni effect due to a gradient of surface tension at the curved surface of the droplets. The induced flow could couple with the macroscopic helix and make it rotate, whence the crucial importance of detecting flows in or out of the droplets in the future. New experiments are in progress to address this issue.

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