1 Introduction

In 1900, Lehmann\(^1\) observed the rotation of the texture of cholesteric droplets submitted to a temperature gradient. A first theoretical explanation was proposed by Leslie in 1968\(^2\) who proved that, for symmetry reasons, the director \(n\) in a cholesteric phase experiences a torque proportional to the temperature gradient \(G\), of expression

\[
\tau_{\text{Leslie}} = \nu n \times (n \times G).
\]

Constant \(\nu\) is called the Leslie thermomechanical coefficient.

In 2008, the Lehmann experiment was reproduced for the first time with a compensated cholesteric liquid crystal (LC)\(^3\) and one year later with a diluted cholesteric mixture.\(^4\) In these two experiments, the cholesteric phase coexists with the isotropic liquid of the LC and forms droplets with a banded texture, which indicates that the cholesteric helix is perpendicular to the temperature gradient. A simple model, based on the existence of the Leslie torque, allowed one to explain the observed relationship between the measured period of rotation, the size of the droplets and the temperature gradient.\(^3\) At this level, it is important to emphasize that this model assumes that there is no flow inside the sample and that the texture rotation is only due to a director rotation induced by \(\tau_{\text{Leslie}}\). From these experiments and the model, values of \(\nu\) were obtained at the melting temperature of the LCs studied. A remarkable point is that the value of \(\nu\) found in this way is proportional to the equilibrium twist of the phase,\(^5\) independently of the concentration of chiral molecules. This last point became obvious when using a compensated mixture in which the compensation temperature coincides with the melting temperature. In that case, the Lehmann rotation disappears in spite of the large concentration of chiral molecules in the mixture.\(^6\)

These results immediately suggested that the Lehmann effect was not due to the Leslie torque, since this torque does not vanish at the compensation temperature as previously shown by observing below the transition temperature the rotation of the director in planar samples treated for sliding anchoring on the two surfaces (Leslie’s geometry).\(^7,8\) This result was reinforced by the fact that the values of \(\nu\) deduced from the rotation of the droplets are completely different (larger by a factor 10 to 1000) from those obtained in the Leslie’s geometry. Even worse, it was shown that the sign of the coefficient could differ between these two techniques. The obvious conclusion is that the coefficient measured from the droplet rotation is not the true Leslie coefficient, but a kind of effective Leslie coefficient, which is now simply called Lehmann coefficient (by opposition to the Leslie thermomechanical coefficient).\(^9\)

In practice, it is also possible to observe droplets with other orientations of the cholesteric helix. For example, Yoshioka et al.\(^9\) observed a coexistence between banded droplets and concentric circle (CC) droplets in which the helix is parallel to the temperature gradient. Similar droplets were also observed in cholesteric mixtures of negative dielectric anisotropy. In these mixtures, CC droplets can be stabilized by applying an AC electric field and can be grown without limit by decreasing the temperature.\(^10,11\) Droplets with the helical axis tilted with respect to the temperature gradient were also observed recently by Yamamoto et al.\(^12\) by using glass plates treated for homeotropic anchoring instead of the sliding planar anchoring.\(^13\) In all these cases, it was also observed that the director was rotating with an

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Do Lehmann cholesteric droplets subjected to a temperature gradient rotate as rigid bodies?\(^†\)

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We performed a Fluorescence Recovery After Photobleaching (FRAP) experiment during the Lehmann rotation of cholesteric droplets in thermodynamic coexistence with the isotropic liquid and subjected to a temperature gradient. By creating and tracking bleached spots near the surface of banded droplets (in which the cholesteric helix is perpendicular to the gradient) and concentric circle droplets oriented by an electric field (in which the helix is parallel to the gradient), we found that neither type of droplet rotates as a solid. This result shows that the texture rotation is mainly due to the local director rotation.
angular velocity much larger than that predicted by the simplified model based on the Leslie explanation.3

In conclusion, there is a large discrepancy between the experiments and the model based on the Leslie explanation. This could mean that the main assumption of the model, namely that there is no flow, is wrong. Indeed, the texture rotation could be explained as well by assuming that the droplets rotate as a rigid body. Such a rotation has already been observed in dye-doped nematic droplets illuminated with a circularly polarized light beam by following the motion of small particles around them.14 In this case, the electric field of the light exerts an external torque on the droplets which is responsible for their rotation. By contrast, the Leslie torque is internal and, for this reason, cannot exert a net torque on the droplet.10 In other words, another mechanism than the Leslie torque must be found for explaining a solid rotation of the droplets, if it exists.

Before starting to look for this new mechanism, it is thus crucial to determine if the droplets rotate as rigid bodies, or if their texture rotation is only due to an individual rotation of the director without flow, as assumed so far in the model. This is the goal of this paper.

This question was already raised by Yoshioka et al.9 who used a photobleaching technique to discriminate between these two types of rotation. In their experiment, the LC is doped with a fluorescent dye. With a laser, a bleached spot is created inside a droplet, the evolution of which is followed in fluorescence microscopy. The existence of a flow is determined by observing the motion of the spot center when the spot is diffusing. Their conclusion was that the banded droplets rotate as a rigid body, whereas in the CC droplets, only the director is rotating (no flow).

Given the importance of these results, we decided to perform again this experiment while changing strategy.

Indeed, one of the main difficulties in the experiment of Yoshioka et al. is to precisely determine the position of the spot center. This is very difficult in banded droplets because the bands remain visible in fluorescence microscopy, which modulates the spot intensity. To avoid this difficulty, we chose to bleach a spot nearby the droplet, in the isotropic liquid, and to follow its time evolution by only analyzing the fluorescence signal outside the droplet. If the droplets are rotating as found by Yoshioka et al., a flow should exist outside the droplet near its surface, and the spot should be advected and stretched by this flow.

In the case of CC droplets on the contrary, our spot was created inside the droplet as Yoshioka et al. did, but we used in our case a liquid crystal of negative dielectric anisotropy with a very small birefringence. This offered us the double advantage of being able to orient the droplets under an electric field while making the internal texture of the droplet almost invisible in fluorescence microscopy.

In addition, we chose cholesteric mixtures in which the droplets rotate very fast in order to be able to follow the spot during at least a half-revolution. This is much more than in the experiment of Yoshioka et al. where the droplets were only rotating by 10–30° during the measurements.

2 Experimental procedure

The two-ovens experimental setup and the procedure used to observe the Lehmann effect have already been described in a previous paper. Briefly, each sample is constituted of two glass plates (covered with an ITO layer when an electric field is required) separated by nickel wires of calibrated diameter. These two plates are covered by a 20 nm thick polymercapran layer, which ensures a planar sliding anchoring. The sample thickness $h$ is measured with a spectrometer to within ±0.1 μm before each experiment. This sample is filled with the studied liquid crystal mixture: 7CB (4,4'-n-heptyl-cyanobiphenyl, Frinton Laboratories) or CCN37 (4z,4'z-propylehexyl-1,1'z-bicyclohexyl-4β-carbonitriles, Nematol), doped with a mass fraction $C$ of the chiral molecule R811 (R-[+]octan-2-yl-4-[4-(hexyloxy)benzoyl]-oxy]benzoate, Merck) and a mass fraction of 0.05 wt% of the fluorescent dye NBD C6-ceramide (N-[2-hydroxy-1-(hydroxymethyl)-3-heptadecenyl]-6-[7-nitro-2,1,3-benzoxadiazol-4-yl]amino, Interchim). This dye is interesting for two reasons: it has a good quantum yield which allows working with very small concentrations of dye and, more important, it is easily bleached by the laser, which avoids to heat the sample and melt the droplet during the laser shot, a problem we met when we used more classical dyes such as the Nile Red or the DiA dyes. Once filled, the sample is then sandwiched between two ovens regulated to within ±1/100 °C. Two very thin layers of glycerol ensure a good thermal contact between the sample and the ovens. It has already been shown3 that the temperature gradient $G$ inside the sample is proportional to both the difference of temperature $\Delta T = T_+ - T_-$ between the two ovens and the ratio of thermal conductivities in the glass and the LC, and depends very little on the sample thickness. This stems from the fact that the layer of LC is very thin compared to the glass plates. In the experiments with the banded droplets (respectively, with the CC droplets), we applied $\Delta T = -10 °C$ (respectively, $-5 °C$), which corresponds to $G = -18 \text{ mK} μ\text{m}^{-1}$ (respectively, $G = -9 \text{ mK} μ\text{m}^{-1}$).

Fig. 1 schematizes the entire setup, constituted of three parts. The middle part is formed by the two ovens and the sample. The upper part is designed to visualize the signal of fluorescence of the sample in reflection. It includes a mercury-vapor lamp illuminating the sample in reflection and a CCD camera (C4742, Hamamatsu) collecting the fluorescence signal through a microscope objective ($\times 10$). The fluorescence filter cube I3 (Leica Microsystems) selects on the input the excitation signal ($\lambda = 450–490 \text{ nm}$) and on the output the fluorescence emission signal only ($\lambda \geq 515 \text{ nm}$).

The lower part is designed to easily create bleached spots in the sample, based on the technique of Fluorescence Recovery After Photobleaching (FRAP). A solid state laser (Sapphire SF 488-100 CW CDRH, Coherent) creates a Gaussian beam (wavelength $\lambda = 488 \text{ nm}$), which is then cleaned by a spatial filter and focused onto the sample thanks to the condenser of the microscope of focal length 1 cm. A custom-built shutter (S1) allows setting the duration of the exposure to the Laser beam, while another shutter (S2) protects the camera during the exposure. In all experiments, the shot duration is 40 ms and the laser power is 10 mW. The shutter S1 can also be replaced
by an absorptive filter which allows observing the spot directly with the camera. In this way, the spot size and its position on the screen of the camera can be finely tuned. The two shutters and the absorptive filter are piloted with an Arduino. The camera and the Arduino are interfaced with a Labview program. Finally, the two ovens and the sample can be moved thanks to two translation stages in order to accurately position the sample with respect to the spot. In addition, a halogen lamp and a semi-reflective plate SR allow us to observe the sample in transmission with natural light. For clarity, the absorptive filter, the polarizers and the translation stages are not represented.

3 Photobleaching in banded droplets

In this section, we present and discuss the results obtained with the droplets, first in a mixture of 7CB doped with 0.05 wt% of the fluorescent dye and $C = 0.25$ wt% of R811 and then in mixtures more concentrated in R811. This first concentration was not chosen randomly but in order to maximize the ratio velocity/size when the droplets have a typical diameter of 50–100 μm. In a sample of thickness $h = 21$ μm, we created a bleached spot near a droplet of radius $R = 43$ μm rotating at angular velocity $ω_⊥ = 0.139$ rad s$^{-1}$. Fig. 2 shows this droplet in fluorescence microscopy just before the shot (photo (a)) and immediately after the spot was created (photo (b)). A first observation is that the background is darker in photo (b) than in photo (a). This is due to the natural photobleaching of the fluorescent dye by the mercury-vapor lamp. A second observation is that this natural photobleaching is not perfectly uniform over the image. This comes from the fact that the lighting is not perfectly uniform too. In practice, we want to only detect the fluorescence signal coming from the spot, and so we need to eliminate all the signal coming from the natural photobleaching and the non-uniformity of the lighting. To do so, we solved the diffusion/advection equation for the dye density by taking into account the destruction term coming from the photobleaching and found an estimate for the only contribution of the bleached spot (renormalized to 1 at time $t = 0$):

$$\hat{s}(x, t) = \frac{I(x, t) - I_0}{I(x, 0) - I_0} e^{a[x(t_0) - x(t)]}.$$  

In this equation, $I(x, t)$ represents the intensity at point $x$ and time $t$ in an image of the acquisition stack, $I_0$ is an average of the additive noise introduced by the camera, and $x$ is the rate of destruction of the dye per unit of light intensity. Time $t = 0$ is chosen just before the laser shot when there is no bleached spot. We refer to the Appendix for the complete demonstration of this formula. Note that, by definition, $\hat{s}$ is approximately equal to 1 far from the bleached spot and is strictly less than 1 inside. For each acquisition, the value of the rate of destruction $x$ is obtained from a linear regression versus time of the quantity

$$\frac{1}{I(x, 0) - I_0} \ln \left[ \frac{I(x, t) - I_0}{I(x, 0) - I_0} \right].$$

Finally, note that the expression of $\hat{s}$ is only valid in the isotropic liquid and not inside the droplet. This stems from the fact that the fluorescence intensity inside the droplet depends on the orientation of the bands, and so would change over time at each point even when there is no photobleaching. This additional phenomenon, certainly due to the variation of the optical index along the helical axis, is not taken into account in our analysis, which only applies for a constant optical index.
For this reason, the spot analysis will be conducted by taking into account only the intensity measured outside the droplet. In the following, we shall display $\hat{s}$ as an image by using an effective $\tilde{s}$ inside the droplet. The latter is obtained by replacing $[f(x,t) - I_0]$ in eqn (2) by a linear regression of the same quantity outside the droplet. This choice is arbitrary but allows us to see the bands and their rotation inside the droplet. This is visible in Fig. 3 where the function $\hat{s}$ corresponding to the droplet shown in Fig. 2 is represented at three different $\Delta t = t - t_{\text{shot}}$ (by denoting by $t_{\text{shot}}$ the time at the end of the laser shot). Note that in our experiment $\Delta t$ is always $\geq 0.5$ s because our homemade camera shutter opens half a second after the end of the shot. We also mention that, for viewing comfort, an adaptive Wiener filter has been used to smooth the images of $\hat{s}$ in Fig. 3. However, all analyses were conducted on the raw signal. These images show that the effect of the natural photobleaching is well corrected by the exponential factor in the expression of $\hat{s}$.

We now turn to the quantitative analysis of these images. Our goal is to determine whether or not the bleached spot is dragged by the droplet when it rotates. A rapid visual examination of the Movie M1 in the ESI† shows that the spot seems to not move.

To confirm this result, we fitted the raw signal $\hat{s}(x,t)$ with the Gaussian function

$$s_g(x, t) = s_0(t) + \Delta s(t) \exp\left\{-\frac{[x - x_0(t)]^2}{2\sigma(t)^2}\right\}.$$  

(4)

The center $x_0(t)$ and the half-variance $\sigma(t)$ were obtained by using a Levenberg–Marquardt regression algorithm. This nonlinear regression was applied by choosing a Region of Interest (ROI) around the bleached spot and by eliminating all points inside both the droplet and the ROI. The remaining points were then processed by the Levenberg–Marquardt algorithm, which allowed us to determine the Gaussian parameters $s_0$, $\Delta s$, $x_0$, and $\sigma$ as functions of time. The two important parameters are the spot center $x_0$ marked by a white dot in Fig. 3 and the half-variance $\sigma/2$ represented by the white circle of equation $|x - x_0| = \sigma/2$ in the same figure. These two elements are also shown in the Movie M1 of the ESL†.

Before commenting these results, we still need to test the validity and the accuracy of the fits. Indeed, a visual check is clearly insufficient, in particular at the end of the recording when the signal becomes very weak. For this reason and for making the comparison between the fit and the raw data more quantitative, we compared two polar marginal distributions calculated from $\hat{s}$ and $s_g$. In the case of $\hat{s}$, these distributions are defined as follows:

$$\hat{g}_r(r, t) = \frac{1}{\pi} \int_0^\pi \hat{s}(r, \theta, t) d\theta,$$

$$\hat{g}_\theta(\theta, t) = \frac{1}{R_{\text{tot}}} \int_0^{R_{\text{tot}}} \hat{s}(r, \theta, t) dr,$$

where the polar referential {r, $\theta$} is centered on the fitted center of the spot $x_0$ at each time $t$. So defined, $\hat{g}_r$ can be interpreted as the mean radial profile inside the upper half-space (where the spot does not overlap with the droplet) while $\hat{g}_\theta$ can be seen as a mean angular profile inside an upper half-disc of radius $R_{\text{tot}}$. In the calculation, the value of $R_{\text{tot}}$ is chosen to be of the same size as the regression ROI, which is always $\geq 3\sigma(t)$. These two integrals are numerically calculated by discretizing the integration intervals and by evaluating $\hat{s}$ at each point with a bilinear interpolation.

In a similar way, the two marginal distributions for the Gaussian model $s_g$ are defined as:

$$g_r(r, t) = \frac{1}{\pi} \int_0^\pi s_g(r, \theta, t) d\theta = s_0(t) + \Delta s(t) \exp\left\{-\frac{r^2}{2\sigma(t)^2}\right\}.$$  

(5)

$$g_\theta(\theta, t) = \frac{1}{R_{\text{tot}}} \int_0^{R_{\text{tot}}} s_g(r, \theta, t) dr,$$

$$\approx s_0(t) + \Delta s(t) \frac{\sigma(t)}{R_{\text{tot}}} \sqrt{\frac{\pi}{2}}.$$  

(6)

The expression of $g_\theta$ follows from $\text{erf}\left[R_{\text{tot}}/(\sqrt{2}\sigma)\right] \approx 1$ as long as $R_{\text{tot}} \geq 3\sigma$. From these definitions, it comes immediately that, in the ideal case of an axisymmetric Gaussian function (as defined in eqn (4)), $\hat{g}_r(r, t)$ is a 1D Gaussian profile which flattens in time, while $\hat{g}_\theta(\theta, t)$ is independent of $\theta$ and decreases as time elapses. By contrast, $g_\theta(\theta, t)$ should depend on $\theta$ if the profile is no longer axisymmetric. This could be the case experimentally, whence the interest of this function to test the axisymmetry of the experimental spot. The interpretation of the functions $\hat{g}_r$ and $g_r$ is still more transparent when the following definitions of the local variance are used:

$$\hat{\sigma}(\theta, t) = R_{\text{tot}} \left[\frac{\hat{g}_\theta(\theta, t) - s_0(t)}{\Delta s(t)}\right] \sqrt{\frac{\pi}{2}},$$  

$$\hat{\sigma}(\theta, t) = R_{\text{tot}} \left[\frac{\hat{g}_\theta(\theta, t) - s_0(t)}{\Delta s(t)}\right] \frac{1}{\sqrt{\pi}} \equiv \sigma(t).$$  

(9)

(10)

These are the functions that we calculated together with the functions $\hat{g}_r$ and $g_r$. They are plotted in Fig. 4. Concerning the experimental radial profile $\hat{g}_r$, we see that, although it is not
perfectly Gaussian at the beginning, it very quickly relaxes towards the fitted Gaussian profile \( g_r \). As for the experimental variance profile \( \sigma_r \), it is quite flat and equal in average to the fitted value of the variance \( \sigma \), even at the end of the recording. This proves that the spot stays axisymmetric. These results validate our method of analysis of the experimental results.

We can now assert that the center of the bleached spot does not move within our precision, in spite of the fact that the droplet rotates one-half turn. More precisely, the spot center has an erratic motion and does not move further than 3.5 \( \mu \text{m} \) from its initial position during the complete acquisition. In addition, and this is crucial in our analysis, the spot remains axisymmetric as shown in Fig. 6 which indicates that there is no visible flow outside the droplet. Our conclusion is that the banded droplets do not rotate as a solid, regardless of the number of bands inside. The contradiction with the conclusion of Yoshioka et al.\(^9\) is perhaps due to an artifact in their measurements that are particularly difficult to interpret when the spot is created inside the droplet.

4 Photobleaching in oriented CC droplets

In this Section, we extend our experiments to the case of CC droplets in which the cholesteric helix is parallel to the temperature gradient. Contrary to Yoshioka et al., we used a LC with a negative dielectric anisotropic, which offers the advantage of being able to orient the helix with an electric field in all the droplets, whatever their size. This allowed us to prepare CC...
droplets much bigger than the size of the laser spot. In addition, we chose a LC with a very small birefringence in order that the CC texture observable in transmission between crossed polarizers becomes almost invisible in fluorescence microscopy. Thanks to this, it is possible to shot inside the droplets and to analyze the bleached spot with the same tools as in the previous section. As for the angular velocity of the helix $\omega_{\Delta t}$, it is measured by observing the droplet in transmission between crossed polarizers just before the shot.

In practice, we used a mixture of CCN37 doped with 0.05 wt% of the fluorescent dye NBD C6-ceramide and $C = 2.6$ wt% of the chiral molecule R811. The birefringence of CCN37 is very small, of the order of $2 \times 10^{-3}$. The reader will note the high concentration of R811 used in this experiment. This is to increase the angular velocity of the texture which we know to be proportional to $C$ in this type of droplets.\(^{10,11}\)

Fig. 7 shows an oriented CC droplet of radius $R = 69 \mu m$ rotating at angular velocity $\omega_{\Delta t} = 0.24$ rad s\(^{-1}\) when $\Delta T = -5^\circ C$. The sample thickness is $h = 22.6 \mu m$. An AC electric field (10 Vrms, 10 kHz) was used to orient this droplet. As the optical indices are almost the same in the cholesteric phase and the isotropic liquid, the droplet is barely visible in fluorescence microscopy. This is the reason why its boundary is marked with a dotted circle in Fig. 7. Photo (a) shows the raw fluorescence image of the droplet just before the shot and photo (b) just after the shot (0.5 s later).

![Fig. 7](image_url) Two raw fluorescence images of a CC droplet coexisting with its isotropic liquid taken before (a) and after (b) a laser shot inside the droplet. An electric field is applied in order to orient the helix parallel to the temperature gradient. The dotted circle marks the boundary of the droplet, barely visible because of the small birefringence of the LC. Mixture of CCN37 + 2.6 wt% R811 + 0.05 wt% NBD C6-ceramide $h = 22.6 \mu m$. The black bar represents 50 \mu m.

Fig. 6 The same as in Fig. 4 for the droplet shown in Fig. 5.

![Fig. 6](image_url)
In order to detect a possible flow inside the droplet, we calculated as a function of time the signal \( \hat{s} \) of the bleached spot inside the droplet, which is possible because of the very small birefringence. This signal is represented as an image in Fig. 8 at five different \( \Delta t \) (see also the Movie M3 of the ESI†). As before, a Wiener filter has been used to smooth the images, but all analyses were conducted on the raw signal \( \hat{s} \). As the calculation of \( \hat{s} \) keeps only the contribution of the bleached spot, the droplet becomes completely invisible in these images. This is why the droplet boundary was emphasized by a black circle. Images of the same droplet taken before the shot in transmission between crossed polarizers and a plot of the intensity measured in the center of the droplet before the shot in transmission between crossed polarizers and a plot of the intensity measured in the center of the droplet were conducted on the raw signal \( \hat{s} \). A stecalculation of \( s^\ast_D \). As before, a Wiener filter has been used to smooth the images, but all analyses were conducted on the raw signal \( \hat{s} \). As the calculation of \( \hat{s} \) keeps only the contribution of the bleached spot, the droplet becomes completely invisible in these images. This is why the droplet boundary was emphasized by a black circle. Images of the same droplet taken before the shot in transmission between crossed polarizers and a plot of the intensity measured in the center of the droplet were conducted on the raw signal \( \hat{s} \). A stecalculation of \( s^\ast_D \).

Finally, we checked again that there was a good agreement between the fit and the data. To this end, we used the variance profiles defined before \( \{\sigma(r,t),\sigma(r,t)\} \) and a slightly modified definition of the radial profiles \( \{g_r(r,t),\hat{g}_r(r,t)\} \):

\[
\hat{g}_r(r,t) = \frac{1}{2\pi} \int_0^{2\pi} \hat{s}(r,\theta,t) d\theta,
\]

where the integration in \( \theta \) is now done over the whole interval \([0,2\pi]\) as the spot lies inside the droplet.

The radial and variance profiles are shown in Fig. 9. Again, we observe that the spot is not perfectly Gaussian just after the shot, but relaxes very quickly towards a Gaussian profile. Although the variance profiles are more noisy than previously and seem to show a slight anisotropy, the standard deviation of \( \sigma \) always corresponds to \( \sim 10\% \) of the mean variance \( \sigma \) at each time, which indicates that the spot globally conserves its shape.

In conclusion, we see that the bleached spot is again well fitted by an axisymmetric Gaussian function till the end of the recording. This analysis and the results shown in Fig. 8 and in the Movie M3 of the ESI† indicate that the spot center does not move significantly (it does not move further than 1.3 \( \mu \)m from its initial position and its trajectory is erratic).

We can thus conclude that there is no measurable flow inside the CC droplets and that the texture rotation is only due to a local rotation of the director. This result is in agreement with the results of Yoshioka et al. We mention that we also checked that there is no flow outside the droplets.

5 Conclusion

The photobleaching experiment shows that there is no visible flow outside the banded droplets as well as inside and outside the CC droplets. These results are in agreement with the results of Yoshioka et al. for the CC droplets. By contrast, the absence of flow outside the banded droplets seems incompatible with the conclusions of Yoshioka et al., namely that the banded droplets rotate as a rigid body (barycentric rotation). We did not try to follow ourselves the evolution of a bleached spot inside a banded drop because the analysis is very complicated. Subtract the band contrast while taking into account the anisotropic diffusion of the dye inside the droplet is indeed very difficult and neglecting these effects could lead to artifacts. A best method could be to follow the motion of an individual dye molecule inside the droplet.\(^{16}\) Such an experiment is planned in the future.

It, of course, remains to understand the physical origin of this director rotation. But one thing is certain: it is not due to the Leslie thermomechanical coupling, of microscopic origin whereas the Lehmann effect is clearly of macroscopic origin as we recalled in the Introduction. The solution could be in the complete resolution of the equations of the nematodynamics (including the thermal diffusion equation for the temperature field), without introduction of the Leslie terms, as L. Kramer suggested before he died in a paper never published.
Appendix: estimation of the bleached spot signal

In this appendix, we show how to process the raw images recorded experimentally in order to extract the only contribution from the bleached spot. Indeed, due to the natural photobleaching by the mercury-vapor lamp, the non-uniform background of the image (typical variation of 5–15% inside the ROI) has a non-trivial time evolution. We represent the recorded images (acquisition stack) by a function \( I(x,t) \), where \( x \) is the position of a point in the image and \( t \) is the acquisition time of the image. As this stack corresponds to a fluorescence signal, we suppose that

\[
I(x,t) = [1 - \chi_{t,[t',t]}(t)] f_{FL}(x)n(x,t) + b(x,t),
\]

where \( f_{FL}(x) \) represents the received light intensity of the lamp (not perfectly uniform) at \( x \), \( n(x,t) \) is the normalized dye density such that \( n(x,0) = 1 \), and \( b(x,t) \) is the additive noise introduced by the camera. Function \( \chi_{t,[t',t]} \) is the indicator function of the time interval \([t',t]\) during which the laser shutter is open and the camera shutter is closed. \( I \) and \( b \) are without unit, but depend on the settings of the camera. In all our experiments, we worked in 12 bits mode, such that the mean and the standard deviation of the noise are respectively \( I_0 \equiv E(b) \approx 201.4 \) and \( std(b) \approx 3.120 \), and the maximum value measurable by the camera is 4096. Here, \( E \) is the mathematical expectation, estimated with an arithmetic mean of a statistical sample, and the standard deviation is defined as \( std(b) = \sqrt{[E(b^2) - E(b)^2]/12} \).

The dye density \( n \) must satisfy the equation of diffusion/advection, with an additional destruction term corresponding to the photobleaching:

\[
\frac{dn}{dt} = D \nabla^2 n - \chi_f n, \quad (14)
\]

where \( d/dt = \partial/\partial t + v \nabla \) is the advective derivative, \( D \) is the diffusion coefficient of the dye in the liquid crystal, \( v \) is the destruction rate of the dye, and \( f_{FL} \) (respectively \( f_{PL} \)) represents the local intensity received from the lamp (respectively from the laser). Looking for a solution of eqn (14) in the form:

\[
n(x,t) = s(x,t)e^{-\int_{t}^{t'}f_{PL}(x)ds}, \quad (15)
\]

yields the following equation for \( s \):

\[
\frac{ds}{dt} - s = D(\nabla^2 s - k^2 s) - \chi_f s, \quad (16)
\]

where \( 1/t = at(\nabla^2 s - k^2 s) \) and \( k^2 = at - \nabla^2 f_{FL} - (at \nabla f_{FL})^2 \).

The value of \( s \) can be obtained from an acquisition without laser shot in an isotropic region, which typically gives \( s \approx 2 \times 10^{-4} \) s\(^{-1} \). In addition, as \( f_{FL}(x) \) represents a nearly - but not perfectly - uniform background, we can assume that \( \nabla f_{FL} \ll \nabla s \). As a consequence, we can neglect in eqn (16) the two terms proportional to \( s/t \) and \( k^2 s \) for a typical experiment duration of 30 s. We can therefore write the final equation for \( s \):

\[
\frac{ds}{dt} = D \nabla^2 s - \chi_f s, \quad (17)
\]

This equation is similar to eqn (14) without \( f_{FL} \). For this reason, it approximates an ideal situation where the lamp would not kill the fluorescence. We can therefore interpret \( s \) as the contribution of the bleached spot to the fluorescence signal and \( \exp(-s f_{PL}(x)t) \) as the contribution of the non-uniform background, slowly decreasing because of the natural photo-bleaching by the mercury-vapor lamp.

Experimentally, we only know \( I(x,t) \) and \( s \). To estimate \( s \), we note that we can write a relation between \( s \) and \( I \):

\[
s(x,t) = \frac{E[I(x,t)] - I_0}{E[I(x,0)] - I_0} \exp[E[I(x,0)] - I_0], \quad (18)
\]

which follows from the fact that \( n(x,0) = 1 \). We then deduce the most simple estimate \( \hat{s} \) of \( s \) by replacing \( E[I] \) by \( I \) (a valid approximation, since the standard deviation of the noise is negligible compared to \( I - I_0 \)):

\[
\hat{s}(x,t) = \frac{I(x,t) - I_0}{I(x,0) - I_0} \exp[I(x,0) - I_0]. \quad (19)
\]

As explained in the body text, this formula is not valid inside the bleached droplets. On the other hand, it applies outside of the bleached droplet and inside (and outside) the CC droplets.

References