

Effect of the electric conductivity on the drift velocity of the cholesteric fingers of the second type in confined geometry

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We show that the drift velocity of the cholesteric fingers of the second species in an ac electric field strongly depends on the conductivity of the liquid crystal chosen and exponentially vanishes above a cutoff frequency we found to be equal (within a numerical factor close to 1) to the charge relaxation frequency of the sample. We further show that the drift velocity is proportional to the applied electric field, provided that the confinement ratio is kept constant. [S1063-651X(99)06409-0]

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I. INTRODUCTION

In 1992, Gilli and Kamayé [1,2] observed for the first time that a polymer cholesteric liquid crystal, confined between two electrodes treated in homeotropic anchoring, may form spirals in an ac electric field. These spirals were then observed in usual cholesteric liquid crystals (i.e., composed of small molecules) by Mitov and Sixou [3,4] and by Ribière *et al.* [5].

These spirals form from cholesteric fingers, which are localized structures that occur at the unwinding transition between the cholesteric phase and the homeotropic nematic phase. This transition is governed by two control parameters: the confinement ratio $C = d/p$ (d is the sample thickness and p is the equilibrium pitch of the cholesteric phase) and the applied voltage between the two electrodes. The former fixes the degree of frustration of the system (we recall that the homeotropic boundary conditions are incompatible with the helical structure of the cholesteric phase), while the latter tends to orient the molecules perpendicularly to the electrodes, provided that the liquid crystal is of positive dielectric anisotropy.

It turns out that the cholesteric-nematic phase transition is first order [6], so that there exists in the parameter plane (V, C) a line $V_2(C)$ where the two phases coexist. The experiment shows that near this line, the cholesteric phase forms elongated structures, called fingers. These fingers can be of different types [7], depending on the conditions of their nucleation. The cholesteric fingers of the first species (CF-1) were known for a long time [8]. They spontaneously form in all samples from the nematic phase. The director field inside a segment of a CF-1 is continuous [9,10] and invariant by a π rotation about the finger axis. On the other hand, its two ends are different [11,12]. As a result, a segment of a CF-1 can crawl along its axis at the coexistence voltage in the ac

electric field [5], but it never drifts perpendicularly to its axis. The situation is different with the fingers of the second species (CF-2). These fingers are more difficult to obtain, except in large- d/p samples (typically larger than 3) in which they form as easily as the CF-1 s [13]. In thinner samples, they can be obtained by stretching cholesteric bubbles [14]. This process shows the close connection between the CF-2 s and the cholesteric bubbles [5]. Furthermore, a segment of a CF-2 has two rounded ends (with a point defect in each of them) that look very much alike in the microscope (although the point defects are not at the same height inside each of them). More importantly, the CF-2 s drift perpendicularly to their axes in an ac electric field, which may lead to the formation of single or twin spirals. From pure symmetry arguments, Gil and Gilli showed that the transverse drift of a CF-2 is due to its noninvariance with respect to a π rotation about its axis. With this in mind, they proposed a new configuration for the CF-2 they called the noninvariant cholesteric finger [15]. Very recently, we proved that this model explains all the static properties of the CF-2 s, as well as their thinning transition above the spinodal limit of the CF-1 s [13].

The problem now is to understand the mechanism responsible for the drift of the CF-2 s in an ac electric field. Gil and Gilly [15] proposed that the Lehman effect [16] or the flexoelectricity [16] could be the motor of the drift. These two models predict the existence of a cutoff frequency above which the drift velocity vanishes. This was indeed observed experimentally [5], but the agreement between theory and experiment is only qualitative.

To go deeper into this question, we systematically measured in samples of different electric conductivities the drift velocity of the CF-2 s as a function of the amplitude and the frequency of the applied voltage. We found that the drift velocity is independent of the conductivity at low frequency. By contrast, it exponentially vanishes above a cutoff frequency close to the charge relaxation frequency of the sample.

The plan of the paper is as follows. In Sec. II, we describe the experiment. In Sec. III, we analyze the effects of the liquid crystal conductivity and of the frequency of the applied voltage on the drift velocity of the CF-2 s. In Sec. IV, we analyze the effects of the amplitude of the electric field in

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the low-frequency regime when the confinement ratio is maintained constant. We conclude in Sec. V.

II. EXPERIMENT

The samples are obtained by mixing the liquid crystal 5CB (4-*n*-pentyl-4'-cyanobiphenyl) with the chiral molecule CB15 (from the Merck Corporation). The cholesteric pitch is measured as a function of the concentration using the Cano-wedge method. All of our measurements in the cholesteric phase are done at 30 °C, i.e., about 3 °C below the clearing point of the mixture. For a concentration of 0.83 wt % of CB15 we found $p = 17.5 \mu\text{m}$. The samples are prepared between two parallel iridium tin oxide (ITO) electrodes treated with silane ZLI3124 from the Merck Corporation to obtain a strong homeotropic anchoring. A square-wave voltage was used. The sample thickness is fixed with Ni wires of calibrated diameters. The sample conductivity is changed by dissolving in the cholesteric mixture a small amount (between 2×10^{-5} and 0.05 wt %) of the ionic impurity tetrabutylammonium bromide (from the Fluka Corporation).

The complex impedance of the samples is measured as a function of the frequency with a precision LCR meter HP4284A (between 20 Hz and 1 MHz). To characterize each sample, we measure its charge relaxation frequency (or Maxwell dielectric frequency [17]) in the isotropic phase (40 °C). Indeed, both the conductivity σ and the dielectric constant ϵ of the liquid crystal are isotropic in the isotropic phase so that the charge relaxation frequency f_{relax} is well defined:

$$f_{\text{relax}} = \frac{\omega_{\text{relax}}}{2\pi} = \frac{\sigma}{2\pi\epsilon_0\epsilon} = \frac{1}{2\pi R_s C_s} \quad (1)$$

where R_s and C_s are, respectively, the resistance and the capacity of the liquid crystal layer. We note that, in the cholesteric phase, the measured value of f_{relax} is of the same order of magnitude (within a factor 2) but may change (within $\pm 20\%$) depending on the cholesteric texture and on the amplitude of the applied voltage.

In order to determine f_{relax} , we measure the complex impedance $R + jX$ of the sample (composed of the liquid crystal layer and of the two ITO layers). An example is given in Fig. 1. In the isotropic liquid, the result does not depend on the amplitude of the applied voltage (which we have varied between 1 mV and 5 V).

In order to fit these data, we use the simple model of Fig. 2. The sample is modeled by a resistance R_{ITO} in series with the liquid crystal layer, which is replaced by a resistance R_s parallel with a capacity C_s . We call V the voltage applied to the sample and V_s the voltage applied to the liquid crystal layer. The former is measured experimentally, whereas the latter must be calculated. Straightforward algebra gives

$$R = R_{\text{ITO}} + \frac{R_s}{1 + R_s^2 C_s^2 \omega^2}, \quad (2a)$$

$$X = - \frac{R_s^2 C_s \omega}{1 + R_s^2 C_s^2 \omega^2}, \quad (2b)$$

and

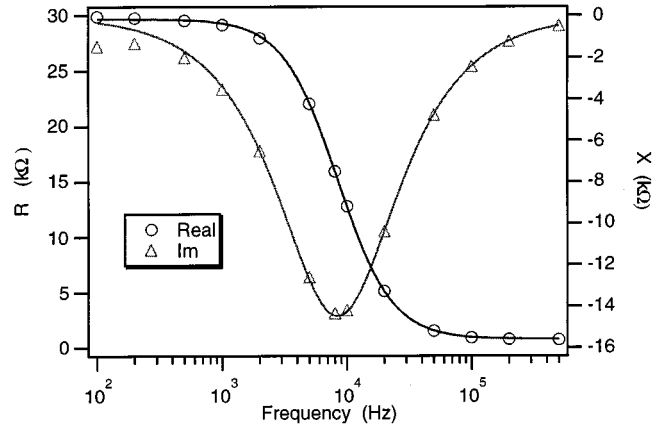


FIG. 1. Real and imaginary parts of the complex impedance of a typical sample in the isotropic phase ($C = 1.7$, $d = 30 \mu\text{m}$, $T = 40 \text{ }^\circ\text{C}$, and $V = 0.1 \text{ V}$). The solid lines are the best fits to Eq. (2) with $R' = 729 \Omega$, $R_s = 28.9 \text{ k}\Omega$, $C_s = 0.65 \text{ nF}$ ($f_{\text{relax}} = 8.4 \text{ kHz}$).

$$V_s = \frac{V}{\sqrt{(1 + R_{\text{ITO}}/R_s)^2 + R_{\text{ITO}}^2 C_s^2 \omega^2}}. \quad (3)$$

We see in Fig. 1 that the fit of the experimental data to Eqs. (2) is good, except at low frequency, for the imaginary part. A way to improve the fit of the two curves is to add a capacity in series with R_{ITO} [18]. This capacity accounts for the formation at low frequency of two Debye layers near the two electrodes. Nevertheless, taking this capacity into account does not change the value of f_{relax} . We also performed all of our experiments at large enough frequencies for this effect to be totally negligible. So we shall forget it in the following.

Finally, we note from Eq. (3) that V is equal to V_s provided that $R_s \gg R_{\text{ITO}}$ and $\omega^2 \ll (1/R_{\text{ITO}} C_s)^2$. These two conditions are fulfilled in our experiments ($V = V_s$ within 5%) so we will not differentiate these two voltages in the following.

III. DRIFT VELOCITY NEAR THE COEXISTENCE VOLTAGE V_2 AS A FUNCTION OF THE FREQUENCY: CONDUCTIVITY EFFECT

In Ref. [5] we already noted that the drift velocity of the CF-2 s strongly decreases above some cutoff frequency, which we did not characterize at this time. We have recently performed new experiments in samples of different conductivities. The confinement ratio chosen is $d/p = 1.7$ with p

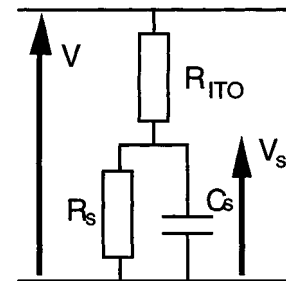


FIG. 2. Schematic representation of the sample. The liquid crystal layer is modeled by a resistance R_s parallel to the capacity C_s . The two ITO layers are replaced by the resistance R_{ITO} .

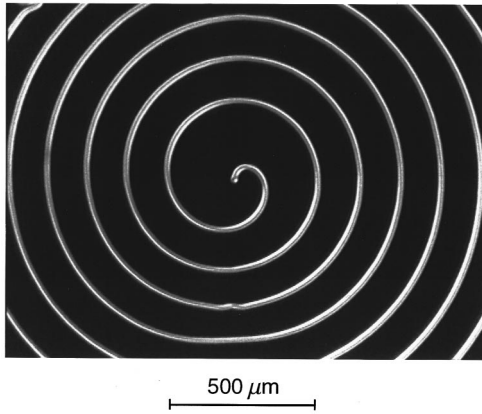


FIG. 3. Spiral observed near the coexistence voltage ($C = 1.77$, $d = 30 \mu\text{m}$, $V = 1.9 \text{ V}$, and $f = 1000 \text{ Hz}$).

$= 17.5 \mu\text{m}$. The drift velocity v_{drift} is measured at a voltage that was typically 0.2 V larger than the coexistence voltage V_2 between the CF-1 s and the nematic phase (in order to avoid finger undulations [5,13]). At this voltage, the CF-2 s bend to form spirals (Fig. 3). It can be easily shown that these spirals are Archimedean far from their centers. In this region, the finger drifts at a constant velocity v_{drift} . We plot in Fig. 4 the drift velocity measured in different samples as a function of the frequency of the applied voltage. For each sample, it is possible to fit $v_{\text{drift}}(f)$ with an exponential law:

$$v_{\text{drift}}(f) = v_0 \exp(-f/f_{\text{drift}}). \quad (4)$$

Two striking results must be noted.

(i) The velocity v_0 in the low-frequency regime does not depend, within experimental errors, on the conductivity of the sample.

(ii) The cutoff frequency f_{drift} is equal to the charge relaxation frequency (within a numeric factor close to 1) (Fig. 5). This last result suggests that the drift mechanism has an electrohydrodynamic origin.

Note that the charge relaxation frequency of our samples is always large, even when we use pure 5CB. This is due to the silane treatment that clearly releases ions in the liquid crystal (without silane and with pure 5CB, we obtain $f_{\text{relax}} \approx 100 \text{ Hz}$).

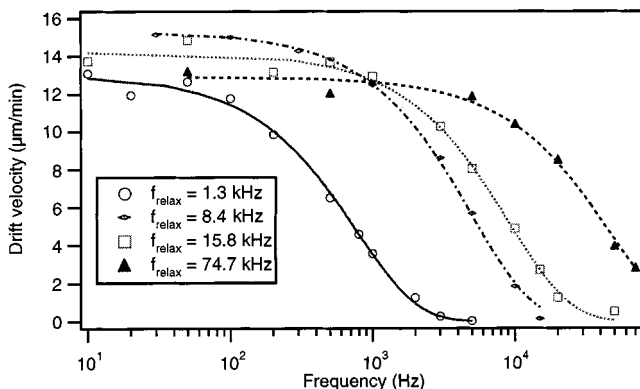


FIG. 4. Drift velocity of the CF-2 s as a function of the frequency of the applied voltage in samples of different conductivities. The lines are the best fits with Eq. (4).

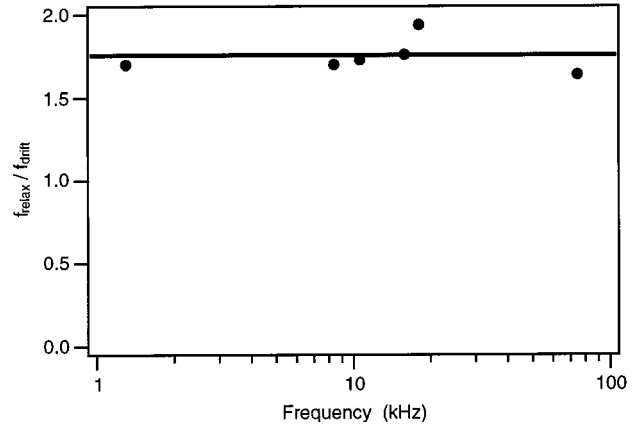


FIG. 5. Cutoff frequency for the CF-2 s as a function of the charge relaxation frequency.

IV. DRIFT VELOCITY AT LOW FREQUENCY AND CONSTANT CONFINEMENT RATIO AS A FUNCTION OF THE AMPLITUDE OF THE ELECTRIC FIELD

We have measured the conductivity-independent low-frequency drift velocity v_0 (at $f \ll f_{\text{relax}}$) as a function of the electric field. Unfortunately, this measurement cannot be done in the same sample because it is impossible to strongly deviate from V_2 . Indeed, the fingers strongly undulate below V_2 , whereas they shorten quickly from their two ends when the voltage is increased above V_2 . As a consequence, spirals are only observable within a small band above V_2 in the (V, C) control parameter plane [5]. To bypass this difficulty, we have made samples of different thicknesses (ranging between 15 and $75 \mu\text{m}$), while maintaining constant the confinement ratio: $C = 1.7$. In this way, the coexistence voltage V_2 , as well as the total energy of the finger, do not change since they only depend on C [12]. By contrast, the average electric field within the sample decreases as $1/d$ as the thickness is increased. In Fig. 6, we plot v_0 as a function of the average electric field $E = V_2/d$. We find that within experimental errors, v_0 is proportional to the electric field.

We conclude that the driving force F_d acting on the finger

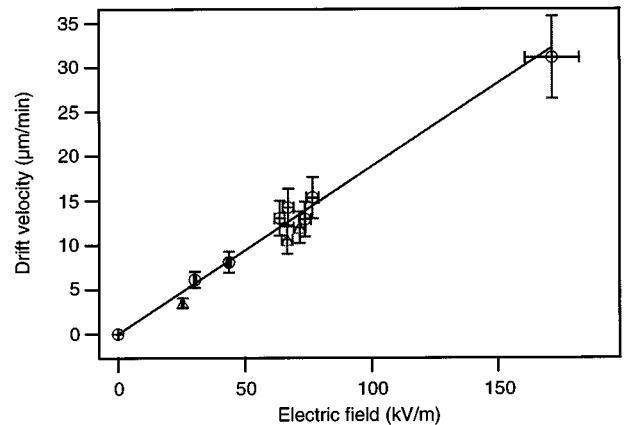


FIG. 6. Low-frequency drift velocity of the CF-2 s as a function of the electric field in samples of different thicknesses but a similar confinement ratio ($C = 1.7$). Two chiral impurities have been used: CB 15 (\circ) and S811 (\triangle).

is proportional to the electric field because the friction force F_v , which opposes F_d (in a steady-state regime $F_d = F_v$), is proportional to v_0 and only depends on C . Indeed, the dissipation Φ can be written in the form [16]

$$\Phi = F_v v_0 = \left[\int \gamma_1 \left(\frac{\partial \mathbf{n}}{\partial x} \right)^2 dS \right] v_0^2, \quad (5)$$

where the integral is taken over a cross section of the finger and γ_1 is a rotational viscosity [16]. Neglecting backflow effects, it can be easily shown that this integral only depends on C .

We have performed a similar experiment with another chiral impurity (S811, from the Merck Corporation, instead of CB15), while keeping the same confinement ratio ($C = 1.7$). We find (Fig. 6) that, within experimental error, v_0 is unchanged. This result suggests that v_0 is independent of the chemical structure of the chiral molecule, which would eliminate a model based on the Lehmann effect, on the condition that the Lehmann coefficient (and thus v_0) depends on the chiral molecule chosen (which is still unclear [19]).

V. CONCLUDING REMARKS

The drift velocity of a CF-2 decreases exponentially and vanishes above a cutoff frequency that is roughly equal to

the charge relaxation frequency of the sample. In the low-frequency regime the drift velocity is independent of the liquid crystal conductivity and, therefore, of the cutoff frequency. It is also independent of the chiral molecule chosen. Finally, the drift velocity is proportional to the electric field at constant confinement ratio.

For the time being, these results cannot be explained by the Gil and Gilli model [15], which does not take into account the conductivity of the sample. In particular, the model based on the Lehmann effect is improbable inasmuch as the drift velocity is independent of the chiral molecule. We think that a model inspired from the Carr-Helfrich mechanism for the convection in the nematic phase must be considered to explain the results (although no convection is observed in the samples). This theory has yet to be proven.

ACKNOWLEDGMENTS

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