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Viscoelastic effects on the core radius of dislocations in cholesterics: a macroscopic coherence length?

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Abstract. — The analysis of the motion of Grandjean-Cano lines in a cholesteric under an applied magnetic field had led to a macroscopic value (1 500 Å) for the core radius $r_H$. A new evaluation, including backflow effects and the dissipation due to the motion of the core, leads to a value of order 0.5 μm. This value is fairly well explained by the limitations of the angular velocity of the director due to viscoelastic relaxation. We thus arrive at the picture of a very anisotropic core extending over a macroscopic distance $r_H$ in a direction perpendicular to the line motion.

1. Introduction. — In a previous paper [1], hereafter referred as (I), the dynamics of the first Grandjean-Cano line in cholesterics under an applied magnetic field has been analysed. As briefly recalled in section 2, this analysis introduced a cut-off radius (around the line) which appeared as a hydrodynamic core radius $r_H$. The value found for $r_H (5,400 \pm 500 \text{Å})$ was surprisingly high, in spite of the fact that a similar value had been found by Geurst et al. [2] in the case of a twisted nematic. This high value raised some criticism. New experiments made at lower magnetic fields gave a smaller value [3] (1 500 Å), but, after a critical look at the assumptions, it has been found that two dissipative processes were neglected in I: i) backflow effects due to the nonuniformity of the magneto-elastic torque and ii) dissipation due to the time-variation of the order parameter in the motion of the core. The contribution of these two processes to the entropy production are evaluated in sections 3 and 4. A new hydrodynamic core radius is derived in section 5. The result is a value ($\sim 10,000 \text{Å}$) still higher than the previous one.
It therefore appears that the hydrodynamic core radius must be very different from the elastic one, which is expected to be of the order of a molecular length. The key lies in a viscoelastic relaxation effect: we show in section 6 that the cholesteric director has an angular velocity \( \omega \) limited by the relation \( \omega \tau_c < 1 \) where \( \tau_c \) is a characteristic relaxation time for cholesterics. A hydrodynamic core radius is derived and found to be quite comparable to the one obtained in section 5.

We conclude section 7 by discussing the physical meaning of this huge hydrodynamic core radius and suggest that it may be related to a dramatic increase in the coherence length due to hydrodynamic effects.

2. Brief recall of the previous analysis. — The first Cano-Grandjean line is a singularity appearing in a cholesteric wedge at a thickness \( d = P_0/4 \) where \( P_0 \) is the cholesteric pitch. Under an applied magnetic field parallel to the wedge, the pitch tends to grow and the line moves. The distortion has been analysed [4] and was shown to extend over a width of order \( d \). When the magnetic field is applied, a local equilibrium is quickly achieved [4] and followed by a slow motion of the line.

This slow line motion was analysed in (I) using energy conservation. The elastic free energy decrease was equated to the entropy production expressed by mean of a dissipation function \( V(d) \) as:

\[
T \sum = \gamma_1 v_L^2 V(d)
\]

where \( v_L \) was the line velocity. This dissipation function was evaluated from experimental results through an effective viscosity \( \gamma_{\text{eff}} \) deduced from a characteristic time \( \tau_0 \):

\[
\tau_0 = \frac{\gamma_{\text{eff}}}{\chi_1 H_1^2}.
\]

\( \tau_0 \) was obtained by fitting the time scale of the theoretical curve to the experimental one.

The value assigned to \( \tau_0 \) in (I) was 95 s. The theoretical analysis developed in (I) is valid only for magnetic fields \( H < H_c \) where \( H_c \) is the critical field for the cholesteric-nematic transition (7.7 kG in our case). The experiments presented in (I) were made at relatively high magnetic fields (5 to 6 kG) and new experiments made at lower fields (3 to 4 kG) lead to the values \( P_0 = 19.7 \mu m \); \( \tau_0 = 127 s \); \( \gamma_{\text{eff}} = 670 \) poise; \( V \approx 1.5 \).

\( V \) was then calculated from the expression:

\[
T \sum_d = \int d^3 r \, \gamma_1 \dot{n}^2;
\]

i.e. for unit length of the line:

\[
= \gamma_1 v_L^2 \int_{-\infty}^{+\infty} dx \int_{-d/2}^{d/2} \left( \frac{\partial \phi}{\partial x} \right)^2 dz
\]

where \( z \) is perpendicular to the plates, and \( x \) is the direction of the line motion. The integral in the r.h.s. of this equation diverges at the origin. A cut-off radius \( \varepsilon \) was therefore introduced and the following expression was obtained, with the assumption \( K_1 = K_3 = K \):

\[
V(d) = \frac{\pi}{4} \left( \frac{K_2}{K} \right)^{1/2} \left[ 1.083 + \ln \left( \frac{K}{K_2} \right)^{1/2} \frac{d}{4 \pi \varepsilon} \right].
\]

Using \( K/K_2 \approx 2 \), \( d = P_0/4 \), one finds

\( \varepsilon \approx 1500 \text{ Å} \).
3. Backflow effects. — In the previous analysis the only dissipative effect came from the director rotation which was assumed to be not coupled to the hydrodynamic backflow. This assumption is correct only if the torque acting on the director is uniform. This is obviously not the case here and we shall now give a crude estimation of the dissipation due to backflow.

Let $\Gamma$ be the non-dissipative torque acting on the director (elastic + magnetic) [5]. In absence of inertial effects $\Gamma$ is exactly counterbalanced by a viscous torque:

$$\Gamma = n \times h = n \times [\gamma_1 (n - \omega \times n) + \gamma_2 A \cdot n] \quad (3.1)$$

where $\omega$ is the angular velocity of the fluid and $A$ the symmetric velocity gradient tensor:

$$\omega = \frac{1}{2} \text{curl } v$$

$$A_{ij} = \frac{1}{2} (\partial v_i / \partial x_j + \partial v_j / \partial x_i).$$

One easily shows [5] that a new force

$$f = \frac{1}{2} \text{curl } \Gamma$$

results from the non uniformity of $\Gamma$. A detailed calculation of backflow effects due to this force involves five viscosity coefficients and is quite impractical. Our purpose here is just to give an order-of-magnitude estimate of the backflow velocities. We thus start from a Navier-Stokes equation:

$$\rho \frac{\partial v}{\partial t} = -\nabla p + \eta \nabla^2 v + \frac{1}{2} \text{curl } \Gamma \quad (3.2)$$

where the pressure has been dropped because it obeys the Laplace equation and thus is uniform. The backflow is a shear flow. The perturbated zone extends again over a distance of order $d$. $\Gamma$ is of order $\gamma_1 \dot{n}$. The characteristic times for rotation of the director and velocity evaluation are of order $d/v_L$ and $v_L$. We first remark that the inertial term is much smaller than the viscous one and can be neglected:

$$\rho \frac{\partial v}{\partial t} \approx \eta \nabla^2 v + \frac{\rho \gamma v_L}{d} \frac{v_L}{d^2}.$$ 

In cgs units, $\rho \approx 1$, $\eta \approx 1$, $v_L \approx 10^{-4}$ cm/s, $d \approx 10^{-3}$ cm, and this ratio is $\approx 10^{-7}$.

The two terms in the r.h.s. of (3.2) must therefore be of the same order:

$$\frac{\eta v}{d^2} \approx \frac{\gamma_1 v_L}{2 d^2}.$$

or $v \approx v_L/2$.

It must here be pointed out that, due to the complexity of nematodynamics, backflow velocities extend along $x$, $y$ and $z$ axes and could be of the same order of magnitude. Taking into account the translational symmetry along the $y$ axis, one has therefore six velocity gradients of order $v_L/2 \ d$. The resulting entropy source is:

$$T \sum_b \approx \eta/2 \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \left( \sum_{i,j} \partial v_i / \partial x_j \right)^2 dx \ dz \ \approx \frac{1}{4} \times \frac{\eta}{2} \times 6 \times v_L^2/4$$

where the factor $1/4$ is obtained by taking for the components of $v$ a sine-like dependence in $x$.  


and \( z \). Replacing \( \eta \) by \( \gamma_1 \),
\[
T \sum_b \approx \frac{3}{16} \gamma_1 v_L^2 \approx 0.2 \times \gamma_1 v_L^2.
\]

We must again emphasize that this estimate is very crude. Velocity gradients were not taken into account in the expression for \( \Gamma \) and all viscosity coefficients were taken equal to \( \gamma_1 \). But it gives a good order-of-magnitude estimate for backflow dissipation.

4. Dissipation in the core motion. — In (I) the core motion had been taken as adiabatic. Let \( r_c \) be the core radius (\( r_c \approx 2 \times 10^{-7} \) cm) and let us compare the characteristic times for the motion of the line (\( \tau_L = r_c / v_L \)) and for heat diffusion (\( \tau_H = \rho C_p r_c^2 / \kappa \)). With \( v_L \approx 10^{-4} \) cm/s, \( C_p \approx 0.2 \) cgs and \( \kappa \approx 3 \times 10^{-4} \) cgs (typical values for organic compounds) one gets
\[
\tau_L \sim 10^{-3} \text{ s} \quad \text{and} \quad \tau_H \approx 3 \times 10^{-11} \text{ s}.
\]
The core motion is therefore not adiabatic but isothermal and some dissipation results.

We shall therefore evaluate the part of the entropy source \( T \sum \) bound to the change of the value of the order parameter \( S \) in the core of the disclination. For this we start from the entropy production due to the change of the tensor-order parameter in the absence of any hydrodynamic flow

\[
T \sum = \int \gamma \dot{Q}_{ij} \dot{Q}_{ij} \, d^3r
\]

where the order parameter \( Q_{ij} \) has components [5]
\[
\dot{Q}_{ij} = \dot{S}(n_i n_j - \frac{1}{3} \delta_{ij}).
\]

Then
\[
\dot{Q}_{ij} = \dot{S}(n_i n_j - \frac{1}{3} \delta_{ij}) + S(n_i \dot{n}_j + n_j \dot{n}_i)
\]

and, using the unit-vector character of \( n \), one easily finds
\[
\dot{Q}_{ij} \dot{Q}_{ij} = \frac{1}{6} \dot{\tilde{S}}^2 + 2 \dot{S}^2 \dot{\hat{n}}_i^2.
\]

Hence
\[
T \sum = \int [\frac{1}{3} \gamma \dot{\tilde{S}}^2 + 2 \gamma \dot{S}^2 \dot{\hat{n}}_i^2] \, d^3r.
\]

In the London approximation (\( S = S_0 \) independent of \( r \)), the entropy production is due only to the rotation of the director. It reduces to \( \int \gamma_1 \dot{\hat{n}}^2 \, d^3r \). Hence
\[
\gamma = (\gamma_1 / 2 S_0^2)
\]

and the entropy production per unit length of the line due to the core motion is
\[
T \sum_c = \frac{\gamma_1}{3 S_0^2} \int_{-d/2}^{+d/2} dz \int_{-\infty}^{+\infty} dx \dot{\tilde{S}}^2.
\]

In order to evaluate this integral we need an analytic expression for \( S(x, z) \). Let us use cylindrical coordinates, calling \( r \) the distance to the line. \( S \) must vanish on the line and \( (S_0 - S) \) must decay as \( \exp(-r/r_c) \). This leads us to use for \( S \) the following expression
\[
S = S_0[1 - 1/cosh(r/r_c)]
\]
which is regular on the line and has the desired asymptotic behaviour. Then

\[ \dot{S} = \frac{\delta S}{\delta x} \frac{dx}{dt} = v_L S_0 \frac{\sinh (r/r_c)}{r_c} \frac{\cos \phi}{\cosh^2 (r/r_c)} \cos \phi , \]

and from equation (4.1):

\[ T \sum_c = \frac{\gamma_1}{3 S_0^2} S_0^3 v_L^2 \int_0^{2\pi} \cos^2 \phi \, d\phi \int_0^\infty \frac{\sinh^2 u}{\cosh^4 u} \, du . \]

A numerical evaluation of the last integral gives 0.3977; one gets:

\[ T \sum_c = \frac{\gamma_1}{3} v_L^2 \times \frac{\pi}{3} \times 0.3977 \approx 0.4 \gamma_1 v_L^2 . \]

This result is independent of \( r_c \), since \( S^2 \) goes as \( 1/r_c^2 \) and the integration volume as \( r_c^2 \).

5. A new evaluation for the hydrodynamic core radius. — Using numerical values, we get from equation (2.4)

\[ T \sum_d = [0.55 \times (1.853 - \ln \{ \alpha r_H \})] \gamma_1 v_L^2 , \]

where \( r_H \) is our new hydrodynamic radius. The total entropy production is now

\[ T \sum = \gamma_1 v_L^2 [0.2 + 0.4 + 0.55(1.083 - \ln \{ \alpha r_H \})] . \]  

(5.1)

The numerical value of \( V \) is deduced from experimental results and therefore not affected by the analysis. Then, using equations (2.1), (2.4) and (5.1), one gets

\[ r_H/\varepsilon \approx \exp(0.6/0.55) \approx 3 \]

and

\[ r_H \approx 0.5 \text{ \mu m} . \]

This estimate is not very precise: crude approximations have been made in section 2. It gives only an order-of-magnitude estimate for the hydrodynamic core radius. Our guess is that

\[ 0.3 \text{ \mu m} < r_H < 1 \text{ \mu m} . \]

6. The hydroelastic relaxation. — The angular velocity of director, \( \omega \), is given by

\[ \omega = \frac{\partial \phi}{\partial x} v_L . \]  

(6.1)

At low angular velocities, i.e. far from the line, the director will follow the molecular field rotation without any problem. However, since \( \partial \phi/\partial x \) diverges on the line, equation (6.1) leads to very high angular velocities, as already remarked by Friedel [6]. In fact \( \omega \) is limited by the condition \( \omega \tau_c < 1 \) where \( \tau_c \) is the viscoelastic relaxation time [5] \( \tau_c = H_2 q^2/\gamma_1 \). Taking \( q_0 = \pi/d = 4 \pi/p_o, K_2 = 3.4 \times 10^{-7} \) dynes and \( \gamma_1 = 2.2 \) poise, one gets \( \tau_c \approx 4 \text{ s} \).

We shall now give an estimate for \( \partial \phi/\partial x \) near the line. \( \partial \phi/\partial x \) is given in (1) as:

\[ \frac{\partial \phi}{\partial x} = - \left( \frac{K_2}{K} \right)^{1/2} \frac{\pi}{2d} \frac{\sin 2 \pi x/d}{\cosh \left[ (K_2/K)^{1/2} 2 \pi x/d \right]} - \cos (2 \pi x/d) . \]
Again taking $K_2/K = 1/2$ and using cylindrical coordinates this expression reduces, for small values of $r$, to

$$\left| \frac{\partial \phi}{\partial x} \right| = \frac{1}{\sqrt{2}} \frac{\sin \phi}{r \left( 1 + \cos^2 \phi \right)}.$$

The average value of the angular factor is $1/2$, and the condition $\omega \tau_c < 1$ now gives

$$r > \frac{v_c \tau_c}{2 \sqrt{2}}.$$

A typical value for $v_c$ is $0.5 \ \mu m/s$ which gives, as a lower limit for $r$, $r_H \approx 0.7 \ \mu m$, in good agreement with the result of section 5.

7. Conclusion. — We have shown in this letter that the surprisingly large core radius found in (1) was related to hydrodynamic effects and a viscoelastic relaxation around the core. The nature and the motion of this core remain open questions. A first approach is to look at the motion of the core as a delayed one. At first sight, this delay should be of order $\tau_c \approx 4 \ s$ and this would lead to a small renormalization of the time scale, i.e. of $\tau_0$.

Let us try to go a little further. For $z \approx r_H$ most of the director rotation occurs outside of the hydrodynamic core. If we assume inside the hydrodynamic core an angular velocity $1/\tau_c$, the delay is very short. On the other hand, for $z \approx r_c$ the rotation of the director is very steep and occurs on a length of order $r_c$. Thus if we keep the picture of two cores, a hydrodynamic one (with radius $r_H$) and an elastic one (with radius $r_c \approx \xi$ of order of a molecular length), we arrive at an upper limit for the elastic core velocity of the order of $r_c/\tau_c \approx 10^{-7} \ cm/s$. This velocity is obviously much too small to allow for the observed motion. We must therefore find some other picture. The locking of the director motion in the hydrodynamic order must result in a strong increase of the elastic stress tensor. In reference [6], Friedel suggested that these large stresses could induce a convective instability inside the core. We would like to make a different (or additional) suggestion. Because of the elastic free energy increase, the balance between the first terms in the Landau free energy:

$$F = \frac{A}{2} S + \frac{C}{3} S^3 + \frac{B}{4} S^4 + \frac{D}{2} (\nabla S)^2$$

and Frank elastic energy is altered and this probably results in a decrease of the order parameter allowing for a faster motion of the director. Note that in a Landau model $\tau_c$ (and then $r_H$) do not depend on $S$. The $S$ decay should occur over a distance of order $r_c$ in the $x$-direction, but could extend over a length $r_H$ along the $z$-axis. One is thus led to the picture of a very anisotropic core where $\xi$ (along the direction of motion) would remain unaltered while $\zeta$ would suffer a dramatic increase under hydrodynamic effects and reach, in our case, a macroscopic value $\zeta \approx r_H \approx 0.5 \ \mu m$. Further experiments could check the soundness of these speculations.

References