Frequency selection of spiral waves in liquid crystals

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We study the frequency selection problem for spiral waves in nematic liquid crystals in an imposed rotating magnetic field. We find that the frequency is selected due to local curvature of the spiral. The results are in excellent agreement with numerical simulations and consistent with experimental data. Possible generalizations to the case of multiarm spirals and anisotropic elastic constants are considered.

PACS number(s): 61.30.Gd, 47.20.Ky

Application of a magnetic field to a nematic liquid crystal leads to the formation of domain walls separating molecules with different orientation [1,2]. These walls, called Brochard-Leger (BL) walls [3], are similar to Bloch and Néel walls for ferromagnets [4]. Rotation of the magnetic field H results in a transition to moving walls and spontaneous formation of rotating spiral waves [5–8]. The core of the spiral wave is an umbilic with the topological charge ± 1 .

A Ginzburg-Landau type equation was derived to describe small distortions of the director field [6,7] in the vicinity of the Frederiks transition [9]. This equation for the single complex order parameter A is of the form

$$\gamma_1 \partial_t A = (\mu + i \gamma_1 \omega) A + \gamma A^* + \frac{K_1 + K_2}{2} \Delta A$$
$$+ \exp(i2\omega t) \frac{K_1 - K_2}{2} \hat{\Delta} A^* - a|A|^2 A, \qquad (1)$$

where γ_1 is the rotational viscosity, μ is the supercriticality parameter, ω is the frequency of rotation of the magnetic field, $K_{1,2}$ are the elastic constants, $\hat{\Delta} = (i\partial_x + \partial_y)^2$, $\gamma \sim H^2$, and a is positive constant (for details see [6,10]). In Ref. [5] the frequency of the spiral rotation and the wave number were measured. Reference [6] reports on the region of existence for spiral waves in the parameter space of Eq. (1). However, such important problems as frequency selection and the character of the interaction with other spirals and boundary remains unclear. In this Rapid Communication we prove that the frequency selection is given purely by the curvature effects of the BL walls far away from the spiral's core. Our result is in excellent agreement with the numerical simulations. The connection with the experimental results of [5] is discussed at the end of the Rapid Communication.

After the scaling $t \rightarrow (\mu/\gamma_1)t$, $\omega \rightarrow \omega \gamma_1/\mu$, $\gamma \rightarrow \gamma/\mu$, $x \rightarrow \sqrt{\mu/[2(K_1+K_2)]}x$, $A \rightarrow \sqrt{a}A$, Eq. (1) can be written in the dimensionless form

$$\partial_t A = (1 + i\omega)A + \gamma A^* + \Delta A - |A|^2 A + \delta K \exp(i2\omega t) \hat{\Delta} A^*,$$
(2)

where $\delta K = (K_1 - K_2)/(K_1 + K_2) < 1$ is the normalized anisotropy parameter. We start from the isotropic case $(\delta K = 0)$; effects of the anisotropy will be discussed later.

We consider a spiral solution in an infinite domain. The spiral frequency selection problem can be obtained from the analysis of the outer solution (far away from the core of the spiral). However, one needs to consider the core of the spiral in order to fix the angular separation between the spiral's arms (interfaces).

In the region of existence of a spiral solution (the parameters $0 < \omega < \gamma < \sqrt{1/9 + \omega^2}$) Eq. (2) possesses two stable equilibria obeying the condition $\sin 2\phi_{1,2} = \omega/\gamma$ [6,11]

$$\phi_1 = \frac{1}{2}\arcsin\frac{\omega}{\gamma}$$
, $\phi_2 = \pi + \frac{1}{2}\arcsin\frac{\omega}{\gamma}$, (3)

$$I_{1,2} = (1 + \gamma^2 - \omega^2)^{1/2},$$
 (4)

where I=|A| and $\phi=\arg A$. Moreover, there exists a one dimensional (1D) front (or kink) solution connecting these equilibria, often called a Bloch wall (BW) or a Brochard-Leger wall [11-13]. The velocity c of the BW in the limit $\omega \ll \gamma$ is given by the expression

$$c = \frac{\pi\omega}{1 - \gamma/3} \left(\frac{(1 - 3\gamma)(1 + \gamma)}{8\gamma} \right)^{1/2}.$$
 (5)

For $\gamma = \omega$ the velocity diverges. For intermediate values of ω , γ the velocity $c(\omega, \gamma)$ can only be obtained numerically.

We assume that the far field of the spiral is represented by two slightly curved BWs. By virtue of the fact that each BW introduces a phase jump π , the 2π periodicity of the phase $\phi(\rho, \theta+2\pi) = \phi(\rho, \theta)+2\pi$ in polar angle θ requires two BWs for each polar radius ρ . For rigidly rotating spiral angular separation α is a constant and does not depend on ρ . However, the angle α between these BWs remains a free parameter and cannot be obtained from the solution of an outer problem. Indeed, if the BWs are far away from each other, mutual interaction is negligibly small and the angular separation of the BW is not fixed. They come close to each other only in the core region. Therefore, matching with the inner (core) solution fixes the angular separation.

Due to the local curvature χ of a BW the normal velocity c_n is modified. To obtain the relation between χ, c, c_n we consider a locally orthogonal curvilinear coordinate system on the interface (BW). The coordinates are the normal λ and the tangential s. Using the fact that in the first order the Laplacian operator is $\Delta = \partial_{\lambda}^2 + \chi \partial_{\lambda} + \cdots$ we obtain the well-known Gibbs-Thomson condition [14,15]

$$c_n = c - \chi. \tag{6}$$

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The condition (6) is fulfilled if the curvature χ remains small. This can be assured if $c \ll 1$, which is the case for $\omega \ll \gamma$ or near the Ising-Bloch transition given by $1-3\sqrt{\gamma^2-\omega^2}\ll 1$.

A rigidly rotating spiral solution is obtained by substituting the expressions for the normal velocity c_n and the curvature χ

$$c_n = \frac{\Omega \rho}{\sqrt{1 + \psi^2}},\tag{7}$$

$$\chi = -\frac{1}{(1+\psi^2)^{3/2}} \frac{d\psi}{d\rho} - \frac{\psi}{\rho\sqrt{1+\psi^2}},\tag{8}$$

into Eq. (6). Here $\psi(\rho) \equiv \rho [d\theta_I(\rho)/d\rho]$, θ_I is the interfacial angle, and Ω is the rotation frequency that has to be determined. One has the following equation for ψ :

$$\frac{d\psi}{d\rho} = \left(\Omega \rho - \frac{\psi}{\rho}\right) (1 + \psi^2) - c(1 + \psi^2)^{3/2}.$$
 (9)

Equation (9) has to be completed by the boundary conditions at $\rho \to 0$ and $\rho \to \infty$. The first condition is that the interface is smooth at the core (planar BW), i.e., $\psi(0) = 0$. The second condition requires the far field to be an Archimedean spiral, i.e., $\psi \sim \rho$ for $\rho \to \infty$.

According to Refs. [14,15], the solution of the problem (9) obeying the boundary conditions exists only if the frequency Ω obeys the relation

$$\frac{c}{\sqrt{\Omega}} = B_0 \approx 1.738.$$
 (10)

Using the expression (5) we obtain immediately the frequency selection formula

$$\Omega = \frac{\pi^2 \omega^2 (1 - 3\gamma)(1 + \gamma)}{8B_0^2 \gamma (1 - \gamma/3)^2} \approx 0.408 \frac{\omega^2 (1 - 3\gamma)(1 + \gamma)}{\gamma (1 - \gamma/3)^2} \,. \tag{11}$$

The formula is considerably simplified in the phase approximation valid for $|\gamma| \le 1$. For this case the phase ϕ becomes an independent variable and is described by a single phase diffusion equation

$$\partial_t \phi = \omega - \gamma \sin 2\phi + \Delta \phi. \tag{12}$$

The frequency in the phase approximation is given by the expression

$$\Omega = \frac{\pi^2 \omega^2}{8B_0^2 \gamma} \approx 0.408 \frac{\omega^2}{\gamma} . \tag{13}$$

The solution of the outer problem provides the frequency selection. However, it is not clear that the outer solution can be matched with the core. Moreover, the angular separation between the BWs remains a free parameter in the outer problem.

Let us return to the core problem. It is important to define the angular separation between the BWs. For $\omega \ll 1$ the core region is roughly defined by the condition $\omega \rho \ll 1$, $\rho \gg 1$. We can construct the core solution by expanding in ω :

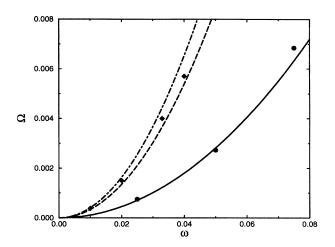


FIG. 1. The selected frequency Ω as a function of ω for γ =0.2 (solid line) and γ =0.1 (dashed line) given by Eq. (11). Diamonds and bullets are the result of numerical simulations with Eq. (2) for δK =0. For comparison, the frequency for γ =0.1 in phase approximation is given by a dot-dashed line [Eq. (13)].

$$\phi = \phi_0 + \omega \phi_1 + \cdots, \quad I = I_0 + \omega I_1 + \cdots$$
 (14)

In zeroth order we obtain the static solution [because $\partial_t = \Omega \partial_\theta \sim O(\omega^2)$]

$$-\gamma \sin 2\phi_0 \Delta\phi_0 + 2\nabla\phi_0 \nabla I_0 / I_0 = 0$$
.

$$I_0 - I_0^3 + \gamma \cos 2\phi_0 I_0 + \Delta I_0 - (\nabla \phi_0)^2 I_0 = 0.$$
 (15)

These equations possess a stable solution containing the topological defect at $\rho \to 0$: $\phi_0 = \theta$, $I_0 \sim \rho$. On the other hand, the far asymptotics of core solution $(\rho \to \infty)$ of Eqs. (15) are represented by two static planar BWs separated by the angle π . One can see this from the symmetry arguments. On the other hand, in the outer solution the curvature vanishes towards the core.

Therefore, matching with the outer solution is possible only if we fix the angular separation between the BWs in the far field to equal exactly π . A spiral with the angle π is called a symmetric spiral [16]. For the symmetric spiral the core is purely flat and the far field solution does not have a cusp singularity at the core (this cusp singularity is typical for any angular separation not equaling π and is smoothed out by diffusion). Clearly, the core possesses a nontrivial topological structure (it includes the topological defect in the spiral's center). However, the frequency selection problem in the first order is not affected by the core.

We performed a numerical solution of Eq. (2). We applied an efficient implicit quasispectral split-step method with noflux boundary conditions. We used typically 256×256 Fourier harmonics. The time step chosen was 0.1 and the size of the integration domain 200×200 . We fixed $\gamma = 0.1, 0.2$ and varied ω in the interval 0.01 - 0.075, which falls into the range of applicability of our theory.

The results of simulations appear to be in convincing agreement with theory (see Fig. 1). Some small discrepancy arises due to numerical problems for $\omega \rightarrow 0$. In that limit the

frequency is very small and the problem becomes a very CPU-intensive project. On the other hand, for larger ω the discrepancy is due to violation of the Gibbs-Thomson condition.

The results presented can be generalized to the case of multiarm spirals described by the equation [8,13]

$$\partial_{i}A = (1 + i\omega)A + \gamma(A^{*})^{n} + \Delta A - |A|^{2}A,$$
 (16)

where n is an integer number. A simple answer can be obtained in the phase approximation. The corresponding phase diffusion equation is of the form

$$\partial_t \phi = \omega - \gamma \sin(n+1) \phi + \Delta \phi. \tag{17}$$

The multiarm solution can be constructed similarly to a two-arm one. The equation $\sin(n+1)\phi = \omega/\gamma$ now possesses n+1 stable equilibria. Therefore the spiral will contain n+1 interfaces (BW) separated by the angle $2\pi/(n+1)$. The frequency can be obtained using simple scaling arguments. Replacing $\tilde{\phi} = (n+1)\phi$, $\tau = t(n+1)\gamma$, $\tilde{\omega} = \omega/\gamma$, $\tilde{x} = \sqrt{(n+1)\gamma}x$ we obtain the equation containing only the parameter $\tilde{\omega}$:

$$\partial_{\tau}\tilde{\phi} = \tilde{\omega} - \sin\tilde{\phi} + \Delta\,\tilde{\phi}.\tag{18}$$

The spiral's frequency $\tilde{\Omega}$ in that scaling is given by

$$\tilde{\Omega} = \frac{\pi^2 \tilde{\omega}^2}{16B_0^2} \,. \tag{19}$$

Coming back to the original variables we obtain the frequency of multiarm spirals:

$$\Omega = \frac{\pi^2 \omega^2 (n+1)}{16B_0^2 \gamma} \ . \tag{20}$$

Let us consider an anisotropy of the elastic coefficients: $K_1 \neq K_2$, which is typical for liquid crystals. The anisotropy results in two effects: oscillations of the BW at the frequency 2ω and a small mismatch of the mean velocity of the BW. In addition, the core of the spiral meanders at the frequency 2ω . Of course, this meandering is completely different from the meandering of spiral waves in isotropic excitable media [14,15] resulting from the intrinsic instability of the core. The correction to the mean velocity is proportional in the first approximation to $(K_1-K_2)^2$ and does not depend on the sign of ω . Hence, clockwise and counterclockwise rotating spirals would have different frequencies, as was observed in numerical simulations [6] and in experiment [2,6]. The experimentally measured anisotropy correction appears to be relatively small even for moderate values of the parameter δK .

The systematic evaluation of the anisotropy correction to the selected frequency is a very tedious problem even in the phase approximation. Here we deduce only the scaling relation between the anisotropy and the frequency up to some constant obtained numerically later on.

The phase equation for the anisotropic case is of the form

$$\partial_t \phi = \omega - \gamma \sin 2\phi + \Delta \phi + \delta K \operatorname{Im}(\exp[2i\omega t - i\phi])$$

$$\times \hat{\Delta} \exp[-i\phi]. \tag{21}$$

We focus on the case $\delta K \ll 1$. We can look then for perturbations of the velocity of the BW. In the first order of δK the velocity undergoes harmonic oscillations at the frequency 2ω and no mismatch of the mean value. Going to the second order we obtain the correction $\sim (\omega \delta K)^2$. The prefactor ω^2 reflects the fact that the anisotropy correction also vanishes with ω . Using scaling arguments we deduce the following expression for the selected frequency:

$$\tilde{\Omega} = \frac{\pi^2 \omega^2}{8B_0^2 \gamma} \pm \beta \, \frac{\omega^3 (\delta K)^2}{\gamma^2} \,, \tag{22}$$

where the constant $\beta \approx 0.4$ has been obtained from simulations. The + sign obtains for clockwise and - for counterclockwise rotating spirals. Indeed, the anisotropy correction $\sim \beta \omega^3 (\delta K/\gamma)^2$ is small (numerically) for $\omega \to 0$ in agreement with experimental data.

We have obtained the unexpected result that the frequency of the spiral is completely defined by the far field and is only slightly affected by the core, similar to the case for spirals in excitable media [14,15]. For spirals in the complex Ginzburg-Landau equation, which is the limit of Eq. (1) for $\gamma \rightarrow 0$ and a complex, the frequency is defined by the core (see, e.g., [17]). Therefore we expect a crossover behavior for small γ and nonzero imaginary part of a. Moreover, by analogy with spirals in excitable media, we expect the velocity of the spiral due to interaction with a boundary or other spirals to fall off superexponentially $(\sim \exp[-\Omega^{3/2}X^3]+\cdots)$ over distances $1 \ll X^2 \ll \gamma^{1/2}/\Omega^{3/2}$ and exponentially weak $(\sim \exp[-\sqrt{2}\gamma X])$ for $X^2 \gg \gamma^{1/2}/\Omega^{3/2}$ (see, for details, [18]). In oscillatory media the spirals interact weakly exponentially [19].

Measurements of the spiral's frequency as a function of ω were performed in Ref. [5]. The measurements were performed far from the Frederiks point, with no electric field. Equation (1) is not formally valid for that case, but may serve as an approximate phenomenological model. A more accurate description requires a more complicated structure of the nonlinear term in Eq. (1) [10]. In Ref. [5] the authors attempted to describe the spiral pattern by a torque equation, coinciding eventually with a phase equation (12). Using the formula (13) we obtained a value of the selected frequency about 2-3 times larger than that experimentally measured. We conclude that the phase approximation is not precise enough, and amplitude corrections should be included. The results can be considerably improved using Eq. (1). The parameters of the equation can be fitted from the experimental data presented in [5]. In particular, one has the value of $\gamma = 1$ and $\gamma_1 = 1/\tau = 1.2048$, where $\tau = 0.83 \text{ s}^{-1}$ is a characteristic relaxation time for the director. From the value $\omega_d \approx 0.87$ one can obtain μ (according to [5] ω_d is the frequency of a dynamic-static transition or an Ising-Bloch transition). For Eq. (1) one has the Ising-Bloch transition for $\mu = 3\sqrt{1 - \omega^2 \tau^2} \approx 2.075$. Moreover, in the experimental data $\omega \tau \approx 1$, and the formula (11) cannot be used as obtained in the limit $\omega \leq 1/\tau$. However, the more general expression (10) is fulfilled fairly well as long as the front velocity c is small.

We compared the experimental data with the selected frequency given by (10) for $\gamma_1 = 1/\tau$, $\mu = 2.075$, $\gamma = 1$. For simplicity we put $\delta K = 0$ (no anisotropy). The front velocity c was obtained from a numerical solution of 1D Eq. (1) for

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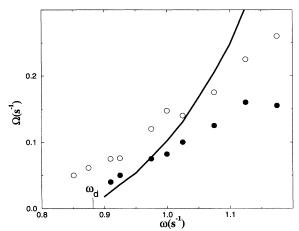


FIG. 2. The selected frequency for μ =2.07, γ =1, γ_1 =1.2048, and δK =0 (solid line) according to Eq. (10). The experimental data [5] are given by open circles (spiral rotating with field) and filled circles (counterfield).

 δK =0. The results are presented in Fig. 2, where one sees reasonable agreement. Deviations from experimentally measured frequencies are caused by anisotropy and violation of the Gibbs-Thomson condition for $\omega \tau \rightarrow 1$. One can also con-

clude that the precise structure of the nonlinear term in Eq. (1) does not affect the frequency. Indeed, in our theory the constant a is scaled away from the beginning, although the structure of the BW is a dependent. For this reason we obtain a reasonable description of the selected frequency using Eq. (1). Presumably, the more complicated nonlinearity [10] can be replaced by the effective nonlinearity $a_{eff}|A|^2A$. Insofar as a_{eff} does not affect the selected frequency, we would have reasonable agreement with the experimental data. Certainly, further improvement of the results can be achieved taking into account anisotropy corrections.

A challenging problem is to evaluate the selected frequency for a liquid crystal in a tilted magnetic field. For that case the system manifests a transition to spatiotemporal chaos above a critical value of the field [20]. The spiral wave for that case is no longer symmetric and the core selects nontrivial angular separation depending on the field magnitude. The envelope equation (1) acquires for this case an additional A-independent term. We expect that our analysis is still valid for this situation. We also expect our result to be relevant for ferromagnets. The model describing the BW in ferromagnets appears to be similar to Eq. (2) (see, e.g., [12]).

This work was supported in part by the Raschi Foundation. Numerous discussions with P. Coullet, T. Frisch, L. Gil, and D. Kessler are acknowledged.

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