# Comment on ''Periodic distortions in lyotropic nematic calamitic liquid crystals''

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Simões Palangana, and Evangelista [Phys. Rev. E **54**, 3765 (1996)] describe the spatial periodicity of director wall arrays in a lyotropic nematic by means of elastic theory. We believe that this treatment is incorrect; the approach cannot describe a wavelength selection mechanism. The texture originates from a dynamic dissipative process. A hydrodynamic description is appropriate and leads to well understood relations between observed wavelengths and viscoelastic parameters. [S1063-651X(97)10211-2]

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The authors of [1] study metastable periodic walls in the director field which appear during the magnetic twist Fréedericksz transition in sandwich cells filled with lyotropic calamitic nematic material. They describe the observed stripe textures in terms of a purely elastic theory. From model functions, elastic energy considerations, and additional *ad hoc* assumptions, relations between the wavelength $\lambda_0$ of the patterns and the magnetic field strength $H$ are developed. The introduction of additional arbitrary elastic parameters is necessary to explain the experimental observations.

During the last two decades, there has been substantial progress in the experimental and theoretical analysis of the magnetically induced dynamic Fréedericksz transition including monomeric thermotropic (e.g., [2–7]) polymeric (e.g., [8–10]) and lyotropic (e.g., [11–21]) materials. The appearance of transient periodic patterns at sufficiently high field strengths (typically $>1.5$ times the Fréedericksz field $H_F$) is a common feature in these systems. The physical origin of the stripe textures was discovered already by Guyon, Meyer, and Salan [2] in the coupling of nonuniform director modes to convective mass flow in the sample. This theory has been applied to the twist Fréedericksz geometry by Lonberg *et al.* [3]. The generation mechanism of the periodic pattern is a well-known hydrodynamic effect. Its description necessarily involves the analysis of the standard hydrodynamic Leslie-Ericksen equations, i.e., the torque balance for the director field and the generalized Navier-Stokes equation. Although elastic terms are involved in these equations, the dominating mechanism is an effective viscosity reduction for nonuniform director reorientation modes. The fastest growing mode determines the periodicity of the resulting director pattern. This hydrodynamic theory has been shown to describe *quantitatively* the stripe pattern orientations, wavelengths, and growth rates in good agreement with the experiment. The magnetic field dependence of the pattern periodicity is well established. The periodic director reorientation results in the formation of metastable wall structures which decay very slowly.

The relation between the wavelengths of these metastable structures and the predictions of the linearized hydrodynamic theory which describes the initial wavelength selection process has been analyzed in a number of publications. It turns out that the wavelength of the fully developed metastable wall pattern is relatively close to the predictions of linear theory [21,5]. The influences of noise [22] and nonlinearities [11] in the hydrodynamic equations has been discussed theoretically. All these investigations prove that the dissipative wavelength selection process is essential for the period of the metastable wall pattern observed in the course of the experiment.

The director structure in the metastable walls has been studied in different systems (see, e.g., [12,17,4,5]). In that situation, elastic theory predicts correctly that the director can escape into the direction normal to the cell plane. The nematic reduces elastic energy by transforming splay bend into twist deformation ($K_{22}<K_{33}$). The elastic description concerns the shape and director structure of the metastable director walls rather than the periodicity of the pattern which is initially determined by hydrodynamics.

The elastic theory as applied by the authors of [1] is not able to describe a wavelength selection mechanism by comparing the elastic energies of deformation modes. It does not even explain the orientation of the wave vector of the pattern. In fact, if we consider a continuous wave vector spectrum there exists an infinite number of periodic equilibrium solutions with different wave vectors. The energetically preferred state, however, is always the homogeneous texture without director gradients in the cell plane. In the experiment, the wall patterns always decay towards this homogeneous solution in finite time (see, e.g., [4]), starting from defects or inhomogeneities in the pattern. The decay time can range from seconds to hours depending upon the viscoelastic properties of the system. Pairs of adjacent walls successively disappear until the uniformly twisted state is reached. There is no finite preferred wavelength from the viewpoint of elastic theory. One must strictly discriminate between the metastable wall textures described here and stable periodic distortions in splay geometry ($H$ normal to the cell) as described in [23] which represent genuine energy minima.

In view of these considerations, the physical model proposed in [1] is therefore not relevant to describe the experimental observations and consequently the conclusions drawn in the paper are incorrect. A detailed discussion of the approach proposed by Simões, Palangana, and Evangelista might nevertheless be instructive. The authors minimize the free energy and derive a closed form of the Euler-Lagrange (EL) equation under certain approximating assumptions. The solution of this equation is not sought analytically. Instead, model functions are introduced which describe a spatially periodic director field. Let us first consider harmonic model functions of wavelength $\lambda$ and amplitude $\varphi_0$, which correspond to the case $l=1$ in the notation of Simões, Palangana, and Evangelista. The model functions are used together with the EL equation to derive relations between magnetic field $H$, wave length $\lambda$, and deformation amplitude $\varphi_0$ (Eqs. (13)–(15) in [1]). The authors misinterpret Eq. (15) as the definition of the selected wavelength $\lambda_0$ which is found inversely proportional to $H^2-H_F^2$ [24]. They ignore the fact that the corresponding amplitude $\varphi_0$ calculated by the insertion of Eq. (15) in Eq. (14) is strictly zero. What is the correct interpretation of Eq. (15) for $l=1$? The result is rather trivial. What the authors can expect from this calculation is to obtain the curves $\lambda(H)$ and $\varphi_0(H)$ for which their model functions are exact solutions of the EL equation. As in the well-known one-dimensional Fréedericksz transition [25], the harmonic functions are exact solutions directly at the critical field. Equation (15) defines the lower wavelength limit $\lambda_C(H)$ for stationary periodic solutions in the given magnetic field $H$. All shorter modes $\lambda<\lambda_C$ are energetically prohibited and their amplitudes decay to zero while for all wavelengths $\lambda>\lambda_C$ energetically stable nonzero solutions can be found.

From the comparison of the misinterpreted Eq. (15) with the experimental data the authors draw the conclusion that modified model functions have to be chosen. An additional parameter $\Delta=(1-l)/2$ is introduced (with $l<1$). The model function consists now of walls of width $\lambda l/2$ separated by regions of width $\lambda\Delta$ with constant deformation $\pm\varphi_0$. Two equations derived from the free energy extremization (Eqs. (14) and (15) in [1]) define $l$ and $\varphi_0$ for given $\lambda$ and thus reduce the number of free parameters to 1. That is, for any $\lambda>\lambda_C$ a set of $\varphi_0(\lambda)$ and $l(\lambda)$ can be found which describes an approximated stationary solution. The comparison of the free energies of these solutions should yield the preferred wavelength provided that such a wavelength exists. Yet after minimizing the free energy with respect to the wavelength $\lambda$ of the test modes, the authors in [1] have a problem. The free energy of the periodically deformed director field continuously decreases when the wavelength $\lambda$ of the deformation is increased. No minimum $F(\lambda)$ exists for finite $\lambda$, and the prediction of a preferred wavelength fails. In order to circumvent this problem, the authors introduce an *ad hoc* term $\alpha\Delta^2$ in the free energy which is proportional to the squared wall distance $\Delta$ and describes an attraction of adjacent walls. The additional free parameter $\alpha$ is then used to fit the experimental data. The introduction of this term is not motivated by the application of elastic theory but its only purpose is to provide an apparent wavelength selection mechanism. This is absolutely unnecessary when one uses the correct dynamical description of the wavelength selection as follows.

After the field $H$ is switched on in the beginning of the experiment, the sample is in an instable equilibrium and the director reorientation is triggered by small fluctuation modes. In a harmonic analysis, $\lambda_C(H)$ (see above) describes the neutral curve in the $(\lambda,H)$ parameter space, which separates the region of stable (decaying) modes from that of the instable (growing) modes. All modes with wavelengths larger than the described critical $\lambda_C$ become instable. Response speed is the decisive criterion for the wavelength selection. The fastest growing mode $Q$ which dominates the resulting pattern periodicity is given by the fourth order equation (see, e.g., [3])

$$h^2=\left(\frac{1-\bar{\alpha}}{\eta\bar{\alpha}}\kappa\right)Q^4+\left(\frac{2\kappa}{\bar{\alpha}}\right)Q^2+\left(1+\frac{\kappa\eta}{\bar{\alpha}}\right), \tag{1}$$

while the normalized wavelength of the neutral curve $Q_C=2d/\lambda_C$ (corresponding to Eq. (15) in [1]) is given by

$$h^2=1+\kappa Q_C^2.$$

We have introduced $Q=2d/\lambda_0$, $h=H/H_F$, and the viscoelastic material parameters $\bar{\alpha}=\alpha_2^2/(\eta_c\gamma_1)$, $\eta=\eta_a/\eta_c$, $\kappa=K_{33}/K_{22}$. One recognizes the critical threshold field $H_C$ for stripe pattern formation:

$$H_C=H_F\sqrt{\left(1+\frac{\kappa\eta}{\bar{\alpha}}\right)}.$$

The data in [1] seem to suggest that the periodic texture appears immediately above the Fréedericksz threshold $H_F$ [see, e.g., Eq. (15)]. In the hydrodynamic description, the preferred sample reorientation is uniform in the cell plane at fields $H_F<H<H_C$. The periodic stripe pattern evolves only above the threshold $H_C$ [3]. In fact, one can confirm the difference between the critical fields $H_F$ and $H_C$ experimentally by means of exact measurements in the vicinity of $H_F$. One also acknowledges from the Eq. (1) that the curve $1/\lambda_0^2$ vs $H^2$ bends from a straight line in accordance with the experimental observations of Simões and others, which is a consequence of the viscosity reduction mechanism.

After the wavelength selection in the hydrodynamic regime, a periodic director pattern has formed and elastic energy is stored in the periodic metastable walls. This energy can be released when the wavelength of the pattern increases. However, the sample cannot simply relax the director deformation by stretching the wall pattern. Pairs of walls have to retreat. This relatively slow process has been studied in several publications before and it is also correctly described in the introduction of [1].

One can prove experimentally that the initial conditions in the reorientation process are essential for the pattern periodicity. When the magnetic field is changed after the formation of the stripe texture, the pattern period is basically determined by the initial magnetic field. In addition to the fundamental discussion above, it should be noted that the assumption of a planar director field (Eq. (1) in [1]) is rather questionable in view of the results published in [12,17,4,5] which give evidence that the director field in the walls escapes towards the cell normal at higher magnetic fields.

In summary, the approach proposed in [1], in particular the introduction of an attractive potential between adjacent walls, is incorrect. The theory is inappropriate for the description of the experimental results reported by the authors. It has to be replaced by the well-established hydrodynamic theory. It should be noted that McClymer, Labes, and Kuzma reported identical experiments with comparable lyotropic samples [21] which the authors of [1] were probably unaware of. In that study, the authors correctly mention the dynamical nature of the wavelength selection process.

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