COMMENTS

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Comment on "Corresponding states of periodic structures in nematic liquid crystals"

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Palangana, Simões, Evangelista, and Arrotéia [Phys. Rev. E **56**, 4282 (1997)] (PSEA) propose a general scaling model for periodic wall patterns in the magnetic twist Fréedericksz transition of nematics, performing an elastic energy analysis. We demonstrate that this model is incorrect because it does not consider consistently the hydrodynamic wavelength selection of the observed structures, but is based on the assumption of inappropriate model functions instead. It is shown that experimental data actually contradict the proposed theory. The approach of PSEA is particularly not suited to determine elastic constant ratios of nematic liquid crystals. [S1063-651X(99)08107-6]

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I. INTRODUCTION

From the thermodynamic description of phase transitions it is well known that physically quite different systems can show universal behavior near critical points, provided the relevant system parameters are properly scaled by their corresponding critical values. It is long acknowledged that the Fréedericksz transition in nematic liquid crystals confined in sandwich cells has much in common with a phase transition [2], and the director deformations in the uniform Fréedericksz transition can be scaled to a universal curve. This is the basis for experimental methods to determine elastic coefficients of nematics (e.g., [3]).

Palangana, Simões, Evangelista, and Arrotéia (PSEA) [1] investigate periodic stripe domains that can be observed in the magnetically induced twist Fréedericksz transition of planar nematic layers, to establish a similar scaling model. PSEA derive equations that describe stationary director deformations in the periodic domains, and introduce *ad hoc* assumptions on the geometrical shape of the director deflections (Eq. (10) in [1]). They claim the discovery of a universal curve of corresponding states. PSEA try to prove that there is a universal relation between the geometrical shape and wavelength of the periodic domains and the magnetic-field strength that can be derived from elastic theory. Finally, they apply their model to their own experimental data of lyotropic nematics and to literature data [4] of thermotropic samples. They suggest that their model is useful for the determination of elastic constant ratios of nematic liquid crystals.

However, the system chosen by PSEA does not belong to the class of phenomena that can be understood within elastic theory, since the formation of periodic domains in the magnetically induced twist reorientation is intrinsically a dynamical phenomenon. In this geometry a planarly oriented nematic layer undergoes the reorientation from a uniform nontwisted initial state to a uniformly twisted final state in the external magnetic field. During this reorientation, one observes the formation and subsequent decay of periodic director patterns, which manifest as parallel stripes in the optical texture. These patterns are relatively long-term persistent on the time scale of the director dynamics, and, therefore, one can use elastic theory to describe the shape (but not the periodicity) of the director field in these structures [5]. The wavelength selection of the patterns is well understood after many authors have devoted intense research to its elucidation (for a short summary see, e.g., [6]). It is established experimentally that the periodicity of walls, which can be observed after a magnetic twist Fréedericksz transition, is selected by a viscosity reduction mechanism during the dynamical reorientation.

The basic inconsistency of the PSEA model is that in the beginning the authors acknowledge the role of hydrodynamic processes in the selection of the pattern wavelength $\lambda(H)$ but subsequently develop their model to discuss the same $\lambda(H)$ curve on the basis of purely elastic theory. However, since the wavelength selection is controlled by hydrodynamics, it is physically obvious that two samples differing only in viscous properties but having identical elastic constants will yield different $\lambda(H)$ curves. Hence the analysis of the periodic structures cannot be reduced to an elastic problem. We emphasize that from elastic theory there are no distinguished stable or metastable equilibrium deformations in the twist Fréedericksz transition other than the in-plane uniformly twisted states. Any preferred wavelength can only be derived from the sample history, which is from hydrodynamics. The scaling of the wavelength vs the magnetic-field curve as done by the authors will only provide information on the initial wavelength selection, which relates λ to H by *hydrodynamic* equations.

A further problem with the PSEA model is that the functions assumed for the nematic director field do not describe metastable but unstable solutions. This will be discussed in the next section. The initial deformations described by PSEA decay rapidly towards the stable uniformly twisted solutions or transform into nonplanar director walls. But even if we assume (like PSEA) that the structures observed can be described by a planar director field, their assumptions on the shape of the director deflections are not justified. This will be shown in the third section of this comment. Since the assumed universal shape parameter is the fundament of the further conclusions of the model, this inaccuracy invalidates the subsequent fits of experimental data.

Finally, PSEA try to demonstrate the existence of corresponding states for lyotropic and low molecular mass thermotropic nematics on the basis of experimental data. An inspection of the respective graph in [1] reveals inaccuracies in the inclusion of literature data. In the fourth section, we reevaluate the experimental curves and supplement some of our own experimental data from literature. We show that the data actually contradict the proposed scaling procedure. The elastic approach presented by PSEA must be rejected as is shown in detail below.

II. CALCULATION OF THE EQUILIBRIUM STATES

PSEA base their model on the solution of the Euler-Lagrange (EL) equation for the free elastic energy. In accordance with the notation in [1], we introduce Cartesian coordinates x,y,z in a planar cell with thickness d, z normal to the cell plane, the planar director field

$$\vec{n}(x,y,z)=[\cos\theta(x,z),\sin\theta(x,z),0],$$

the twist Fréedericksz field $H_F=(\pi/d)\sqrt{K_{22}/\mu_0\Delta\chi}$ [8], and normalized quantities

$$h=\frac{H}{H_F},\quad t=\sqrt{\frac{K_{22}}{K_{33}}}\frac{\pi}{d}x,\quad \tau=\sqrt{\frac{K_{22}}{K_{33}}}\frac{\pi}{d}\lambda.$$

The solutions of the EL equation in the limit of small deformations $\theta\approx\eta(t)\cos(\pi z/d)$ and elastic two-constant approximation ($K_{11}=K_{33}$) can be found from

$$C=\frac{1}{2}(\partial_t\eta)^2+\frac{1}{2}(h^2-1)\eta^2-\frac{1}{4\theta_0^2}h^2\eta^4,\tag{1}$$

where $\theta_0^2=8/3$ according to PSEA; a more correct approximation yields $\theta_0^2=2$ [7]. We note that the small angle approximation is certainly not justified for the description of the patterns observed in PSEA's experiments, cf. the calculations in [5]. For $h>1$, this equation has the trivial solution $\eta\equiv 0$ and the two uniform solutions $\eta=\pm\eta_\infty=\pm\theta_0\sqrt{h^2-1}/h$. The first one is instable and the two latter are asymptotically stable. In addition, the system has an infinite number of periodic solutions, which can be found when Eq. (1) is treated as a periodic boundary value problem with $\eta(i\,\tau/2)=0$ $(i=0,\pm1,\pm2\dots)$. When such a periodic solution is regarded, the constant C is related to the derivative of the deflection angle at the nodes $t_{i0}=i\,\tau/2$ of $\eta(t)$ at the domain boundaries:

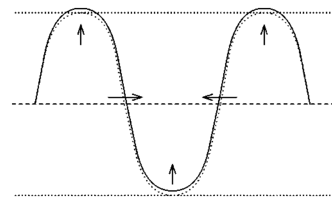

FIG. 1. Sketch of the deflection $\eta(t)$ of the director field. Dots mark the equilibrium solution. The domain in the middle with a slightly lower amplitude is extinguished finally.

$$C=\tfrac{1}{2}(\partial_t\eta)^2|_{t=t_{i0}},$$

and in the same way to the maximum deflection angle η_0 in the domains

$$C=\frac{1}{2}(h^2-1)\eta_0^2-\frac{1}{4\theta_0^2}h^2\eta_0^4.$$

The solution has the form of an elliptic sine function [1]

$$\eta(t)=\eta_0 sn\left[\sqrt{(2\eta_\infty^2-\eta_0^2)\frac{h^2}{2\theta_0^2}}\;t,k\right].$$

The shape parameter k in [1] can be expressed by the amplitude $\eta_0(\tau,h)$ of the periodic deformation with normalized wavelength τ and that of the uniform deformation in the form $k^2(\tau,h)=\eta_0^2/(2\eta_\infty^2-\eta_0^2)$. Of the five parameters h,τ,C,k, and η_0, only two can be chosen independently in the elastic description (Eqs. (9) ff in [1]). If, for example, τ and h are given, one can determine k from

$$\sqrt{1+k^2}K(k)=\tfrac{1}{4}\;\sqrt{(h^2-1)}\;\;\tau\tag{2}$$

[Eq. (9) of PSEA], where $K(k)$ is the complete elliptic integral of first kind.

The solution given by PSEA describes (for $h>1$) the stable deformation in a box cell with extensions $\lambda/2$ along x and d along z under fixed boundary conditions $\theta=0$ at the walls. However, the assumption that a periodic sequence of such solutions with alternating sign of θ describes a *metastable* array of splay-bend (SB) walls would be incorrect. Actually such a deformation is stable with respect to distortions of θ with the same periodicity λ along x, but is instable to long-wavelength fluctuations. Mathematically, it has been demonstrated recently by Amengual *et al.* [9] that all the oscillating solutions are linearly unstable with respect to a uniform perturbation. Figure 1 visualizes the situation. Consider the boundary t_{i0} between two adjacent domains $i,i+1$, with $\eta(t_{i0})=0$. An infinitesimal fluctuation of η, which shifts this zero in one direction, leads to the growth of the deflection amplitude in the larger domain and the decrease of the amplitude in the other; the resulting torque on the director field increases the drift of t_{i0}. The larger domain expands on the expense of the smaller one until the latter is extinguished. This instability leads to the continuous decay of the pattern by disappearance of individual domains, even if other effects are disregarded. A statement about the stability of the calculated solutions was not directly made in [1], but since

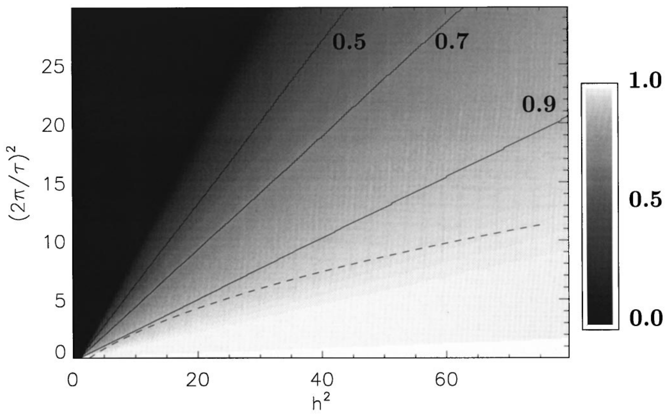

FIG. 2. The profile of $k(\tau,h)$ in the two-dimensional parameter space. In the black area stationary periodic solutions are forbidden. The gray scale maps different values of k. The straight lines visualize three states of equal k (i.e., equal τ/τ_c). The dashed line corresponds to the $\tau(h)$ dependence of Lonberg's experiment [4].

the authors discuss equilibrium states it is important to note that their approach only describes unstable deformations of the director field.

In addition to this instability, which leaves a uniformly twisted state $\eta=\pm\eta_\infty$, SB walls may also become unstable with respect to an out-of-plane escape of the director [5,10–13]. When one observes long-term persistent patterns in the experiment described here, one usually deals with nonplanar escaped twist-bend (TB) walls. These are commonly considered metastable from experimental evidence, although a mathematical analysis of their stability still lacks.

III. SHAPE OF THE WALLS

Even if we suppose planar stationary SB walls, the correct way to describe their shape should start with the calculation of $\tau(h)$, from the linear stability analysis of fluctuation modes during the reorientation [4]

$$h^2=\left(\frac{1-\overline{\alpha}}{\overline{\eta}\overline{\alpha}\kappa}\right)\left(\frac{2\pi}{\tau}\right)^4+\left(\frac{2}{\overline{\alpha}}\right)\left(\frac{2\pi}{\tau}\right)^2+\left(1+\frac{\kappa\overline{\eta}}{\overline{\alpha}}\right), \quad (3)$$

with $\kappa=K_{33}/K_{22}$, and $\overline{\alpha},\overline{\eta}$ are viscous parameters of the nematic. For given h, one can determine τ from Eq. (3) and insert it in Eq. (2) to calculate $k(h)$.

The role of this dynamic wavelength selection is mentioned by PSEA in their introduction of the system but subsequently disregarded. Instead, the new idea in the PSEA model is the introduction of a guess for the parameter k, which controls the shape of the director walls:

$$k(h)=1-e^{-\alpha(h-1)} \quad (4)$$

(Eq. (11) in [1]). The authors claim that the parameter α is universal for nematics. PSEA derive this simple relation from arguments, which may be plausible for structures of a *fixed* wavelength and a variable magnetic field. However, the assumption that the wavelength is independent of h contradicts the experiment.

We demonstrate by an example that the proposed function $k(h)$ is inadequate. Figure 2 visualizes the general function $k(\tau,h)$ calculated from Eq. (2) in a two-dimensional parameter space. The parameter k is constant for equal τ/τ_c where $\tau_c=2\pi/\sqrt{h^2-1}$ is the wavelength of the neutral curve, that is, the lower boundary of τ for energetically allowed stationary periodic deformations at given h. The actually selected wavelength derived from Eq. (3), with the viscoelastic parameters of the liquid crystal MBBA from [4], is indicated by the dashed line in Fig. 2. Along that line, the relation

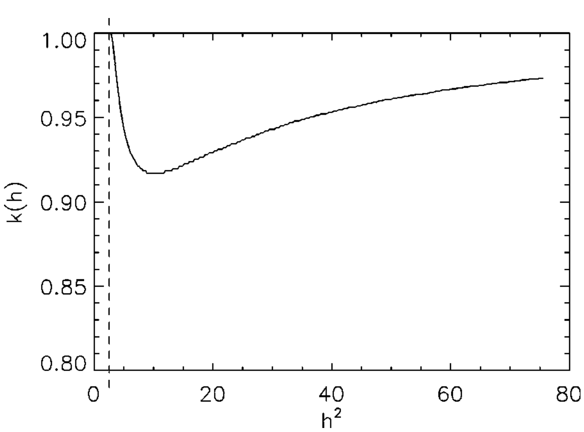

FIG. 3. The $k(h)$ curve derived with $\tau(h)$ taken from the linear stability analysis. Parameters of Lonberg *et al.* [4] for MBBA have been assumed to obtain $\tau(h)$. The dashed line depicts the critical field for periodic pattern formation h_c.

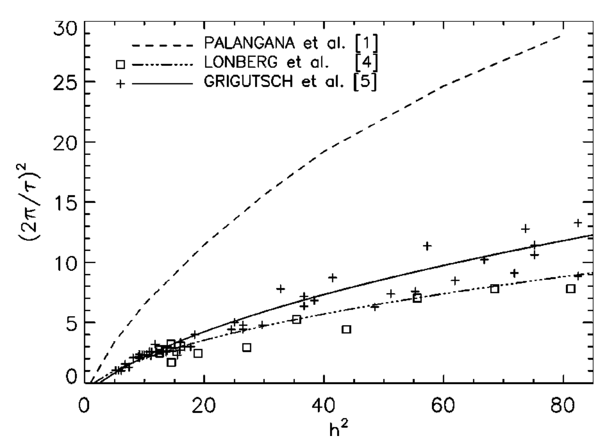

FIG. 4. Comparison of the curve given by PSEA [1] with experimental data of Lonberg *et al.* [4], and Grigutsch *et al.* [5]. More detailed data in the vicinity of the critical field can be found in [13]. The curves demonstrate clearly that the scaling model of PSEA fails.

between k and h for stationary domains can be calculated by means of Eq. (2), and we present the result in Fig. 3. Although this curve is not universal but in details dependent upon viscoelastic properties of the material, its general shape is representative, and it is qualitatively different from the curve assumed by PSEA with Eq. (2). In particular, periodic patterns set in only above some well defined threshold $h_c = \sqrt{1+\kappa\overline{\eta}/\overline{\alpha}}>1$ defined by Eq. (3), with $k\approx 1$ near the threshold field.

IV. COMPARISON OF EXPERIMENTAL DATA

PSEA compare experimental data in their Fig. 2, suggesting that there is one fundamental master curve for different nematic media. However, the authors obviously have made an error in the scaling of Lonberg's data. We present the reevaluated curves in Fig. 4, the correct substitution is $(K_{33}/K_{22})\ (2d/\lambda)^2=(2\pi/\tau)^2$. Open squares and the corresponding dash-dotted fit curve denote MBBA data of Lonberg *et al.* [4], crosses represent our data for a thermotropic mixture [5] together with the solid fit curve, and the dashed line gives the graph presented by PSEA for their lyotropic samples. These scaled curves are far from being identical. We note that in our experiments, K_{22}, K_{33}, and $\Delta\chi$ have been determined in independent measurements. The elastic constants and $\Delta\chi$ for MBBA in Lonberg's experiment are also known from literature. In contrast, PSEA have treated K_{22}, K_{33} for their substances as *fit* parameters, which have not been compared to independent experiments. This gives additional freedom to scale the curves and, therefore, it is understandable that the three graphs for lyotropic materials coincide.

It is important that in the vicinity of the critical fields h_c for pattern formation (which do not coincide for all samples), the curves on principle cannot be brought to coincidence with the scaling proposed. We underline that the dynamical twist Fréedericksz transition is uniform for $1<h<h_c$ (see, e.g., [6]), that is, the model of PSEA fails in particular in the vicinity of the threshold field.

Since the curves presented by PSEA start at $h=1$, it is highly probable that the authors have not determined the Fréedericksz field H_F correctly, but do not distinguish between the fields H_F and $H_C=h_c H_F$ and scale their magnetic field with H_C. This is an additional error source when elastic data are retrieved from the wavelength curve.

V. SUMMARY

In our opinion the paper by PSEA is based on an incorrect assumption on the type of their periodic patterns. Due to their anisotropic and nonlinear physical properties, nematic liquid crystals provide many examples for spontaneous pattern-formation processes; among the different types of periodic patterns are thermodynamically stable or metastable periodic equilibrium states (e.g., in the splay Fréedericksz transition [14]), dissipative structures (e.g., electroconvective rolls [15]), and transient patterns (e.g., in the magnetic Fréedericksz transition, see [4]). Despite many similarities in their optical appearance, these systems have completely different physical origin. Only the first type can be understood within elastic theory.

Palangana *et al.* have attempted to develop a purely elastic model for stripe patterns formed during the magnetically induced reorientation of nematics, which are intrinsically related to the third type of patterns. Therefore, the proposed model is not sufficient to describe the system. The presented theory of corresponding states in the twist Fréedericksz transition of nematics is not correct. In particular, the approach cannot be used to determine elastic constant ratios.

In the Introduction, the authors repeat an incorrect statement about an alledgedly existing attractive potential between nematic director walls. This has been disproven in a previous publication [6].

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