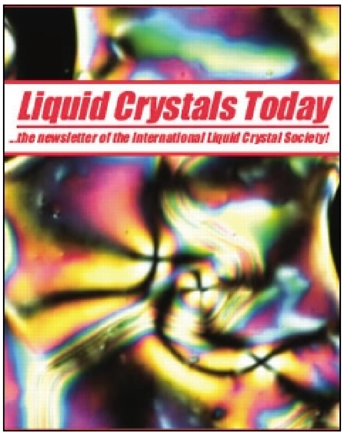

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Scientific Note

O. D. Lavrentovich[a]; P. Palffy-muhoray[a]
[a] Liquid Crystal Institute and Chemical Physics Program, Kent State University, Kent, OH, USA

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Scientific Note

Nematic Structures in Composite Materials

O.D.Lavrentovich and P. Palffy-Muhoray

Liquid Crystal Institute and Chemical Physics Program, Kent State University, Kent, OH 44242, USA

Composite materials containing nematic liquid crystals (such as liquid crystals in porous matrices [1–5], polymer-stabilized liquid crystals [6,7], liquid crystals filled with small solid particles [8,9] are fascinating physical systems which have generated considerable current interest. The purpose of this note is to point out a simple but curious feature of nematic liquid crystals which governs the basic structural properties of these composite systems, yet one which, in our opinion, has not received sufficient attention in the literature.

Since liquid crystal samples are necessarily surrounded by boundary surfaces, their free energy consists of both bulk and surface contributions. For example, in a domain with linear dimension R, the bulk elastic energy associated with director deformations is $F_b \cong \int \frac{1}{2} K (\nabla.\hat{n})^2 dV \sim KR$, while for comparable surface distortions, the surface energy is $F_s \sim WR^2$. The ratio of these is $F_s/F_b \sim R/l_e$, where K is an elastic constant, W is the surface anchoring strength, and $l_e = K/W$ is the de Gennes-Kléman extrapolation length: typically, $l_e \approx 0.1 - 10\,\mu m$. The relation $F_s/F_b \sim R/l_e$ gives rise to the feature that *in large domains surface interactions dominate, while in small domains bulk elastic effects dominate*. This behaviour is opposite to what is usually observed in conventional materials, and stems from the fact that the director deformations are Goldstone modes, that is, the energy density associated with deformations with wavelength R is K/R^2.

In composite systems with randomly oriented interfaces between the liquid crystal and the rigid matrix (skeleton of a porous medium, suspended solid particles, rigid polymeric rods, etc.), connectivity of the nematic subphase and the competition between bulk and surface contributions lead to the following result. If the length scale L of the disturbance (pore size, radius of polymer thread, etc.) is larger than the extrapolation length,l_e., then the director field, $\hat{\mathbf{n}}(\mathbf{r})$ is spatially non-uniform, and in general $<\hat{\mathbf{n}}(\mathbf{r})>$sample = 0. By contrast, when L is smaller than l_e, then $\hat{\mathbf{n}}(\mathbf{r})$ = *const.* and the director field may be uniform over lengths much greater than l_e.

Typically, the structure of the medium in contact with the liquid crystal defines an effective length scale L (e.g., the size of pores, diameter of the suspended particle). Generally, $L \neq l_e$, but we assume both to be larger than the nematic correlation length to maintain the validity of the director field approach. If $L >> l_e$, then $KL << WL^2$ and the equilibrium configuration is the one which minimizes the surface anchoring energy. The resulting director configuration may be highly distorted, with $\hat{\mathbf{n}}(\mathbf{r})$ following the changing directions of anchoring forces at the curved interfaces, and generally $<\hat{\mathbf{n}}(\mathbf{r})>_{sample} = 0$. If $L << l_e$, then $KL >> WL^2$ and the equilibrium configuration is the one which minimizes the bulk elastic energy F_b. Since the nematic subsystem is connected, this implies that $\hat{\mathbf{n}}(\mathbf{r})$ = *const.* is favoured, and thus the director may be uniform over long length scales – possibly everywhere in a finite sample.

To illustrate the above, we consider the example of an 'H'-junction, formed by cylindrical pores of radius r and length L ($L \geq r$). The walls of the pores favour planar alignment; that is, alignment with the director parallel to the axes of the cylinders. There are two limiting cases: (a) the director field $\hat{\mathbf{n}}(\mathbf{r})$ minimizes the surface energy everywhere in the pores and consequently defects form near the junctions (Figure 1(a)), or (b) the director field $\hat{\mathbf{n}}(\mathbf{r})$ = *const.* minimizes the bulk energy everywhere, and thus there is a surface energy cost, at least on some of the walls (see bold lines in Figure 1(b)). The excess energy of configuration (a) is the elastic energy at the junctions, $F_b \sim Kr$, while that of configuration (b) is the surface energy of the central pore due to unfavourable alignment, $F_s \sim WLr$. The ratio of these energies is again $F_s/F_b \sim R/l_e$, and a 'monocrystal' appears to be energetically favourable when $L << K/W$. Similar conclusions can be drawn for pores with different boundary conditions.

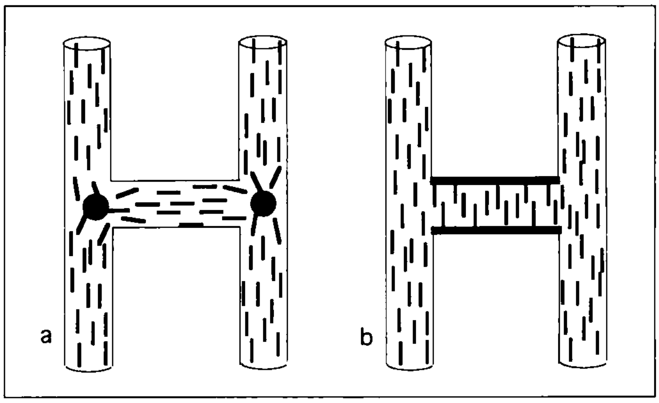

Figure 1 *Equilibrium director configurations in 'H'-junction of pores. Configuration (a) is stable if the pore size L is greater than the extrapolation length l_e; configuration (b) is stable if L << l_e.*

We next consider a nematic matrix in which solid spheres with radius r are dispersed. The preferred alignment is assumed to be homeotropic; that is, the surface energy is minimized when the director is normal to the interface. We are interested in how the configuration of the system depends on the sphere radius r and the volume fraction ϕ. In the homeotropic state, the elastic energy is $F_b \sim NKL$, where L is the mean distance between the N spheres, while in the uniform $\hat{\mathbf{n}}(\mathbf{r})$ = *const.* state, the surface energy is $F_s \sim NWr^2$. Since $L \approx \phi^{-1/3}r$, $F_s/F_b = \phi^{1/3}r/l_e$, and the uniform $\hat{\mathbf{n}}(\mathbf{r})$ = *const.* state is expected when $r < \phi^{-1/3}l_e$, or $L < \phi^{-2/3}l_e$. Although the total interfacial area increases when ϕ = *const.* and $r \rightarrow 0$, the influence of anchoring decreases.

A similar situation occurs for polymer stabilized materials. We consider a nematic containing a random network of polymer chains which subdivided the system into N cells of size L (length of a rigid polymer segment); the radius of the polymer rod is taken to be r. For the distorted state, the bulk energy is $F_b \sim NKL$, while the surface energy of the uniform state is $F_s \sim NWrL$. Since $r \approx \phi^{-1/2}L$, $F_s/F_b \sim \phi^{1/2}L/l_e$, one finds a critical mesh size $L = \phi^{-1/2}l_e$ below which a uniform director field $\hat{\mathbf{n}}(\mathbf{r}) = const.$ appears favoured.

We point out, however, that even for very small mesh sizes L, we do not expect true long range uniform alignment. In accordance with the random field arguments of Imry and Ma [10], long range order is destroyed by random fields such as surface interactions in dimensions $d \leq 4$. In our case, this comes about because long wavelength distortions of the director field have a vanishingly small energy cost: these are therefore still imposed by the random medium. We argue, however, that in systems whose characteristic length L is smaller than the extrapolation length l_e, large nematic domains with nearly uniform alignment can form. The domain size R may be comparable to the sample size, and we suppose R to be of the form:

$$\frac{R}{l_e} \approx \frac{l_e}{L} + \frac{L}{l_e}$$

Our conclusions about the existence of uniform macroscopic structures do not mean only that weak anchoring cannot compete with the elastic forces and the individual nematic subvolumes are uniform. We argue the more subtle point: for systems with characteristic lengths smaller than the extrapolation length., the connectivity of the nematic volume makes the entire system uniform, since the elastic energy saved is greater than the penalty for surface misalignment.

A number of experiments suggest themselves to verify the above arguments. The characteristic size R of liquid crystal domains in heterogeneous materials should behave non-monotonically with the effective length scale L of the heterogeneity; $R \sim L$ for $L > l_e$, but diverges for $L << l_e$, until it reaches the sample size. In this latter 'monocrystal' state, the system should exhibit long correlation lengths and relaxation times, and increased sensitivity to orienting external fields. Indications of such behaviour may already have been observed in recent experiments [2,5].

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Review of

Liquid Crystals Topics in Physical Chemistry

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by J.W. Emsley, Department of Chemistry, University of Southampton

One of the joys of doing research on liquid crystals is the interdisciplinary nature of the subject, but this also makes it difficult for those new to the topic. Introductory texts have a hard task since they should include something about chemical structure and structure-property relationships, something on the macroscopic physics, and perhaps too, something on electro-optic devices. And all this has to be digestible to scientists and engineers with diverse backgrounds. This collection of articles assembled by Stegemeyer is a welcome attempt to satisfy these constraints. It comprises six chapters, each by a different author. Demus introduces the polymorphism of liquid crystals and gives a detailed description of most of the thermotropic phases and discusses the influence of chemical structure on phase stability. Petzl deals with the macroscopic physics, such as the elastic constants, viscosity, and the effect of applying electric and magnetic fields. Zentel describes liquid crystal polymers, and Hiltrop deals with lyotropic systems. Pohl describes some of the uses of liquid crystals as solvents in spectroscopy, and finally Schadt describes liquid crystal displays.

The aim of an introductory text should be to introduce the reader to a body of knowledge, and to the main principles which are invoked to explain observations. From this point of view, I found the articles by Zentel and Schadt to be excellent, and in particular they both bring the reader fully up to date. Of the other chapters, only that by Pohl seriously disappoints. It is different in kind from the rest of the book, being concerned with how using a liquid crystal as a solvent can be exploited in spectroscopy, rather than how spectroscopy can be used to study liquid crystals.

In Southampton we offer a course on liquid crystals to our final year undergraduates, and in the absence of a suitable introductory text, we give a wide reading list. This book will certainly feature high on this list as being perhaps the best yet available.