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## Morphology Development in Liquid-Crystal/Polymer Mixtures

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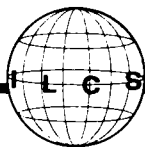
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# Liquid Crystals

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**TODAY**

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## EDITORIAL

### Liquid Crystals Today

Members of the International Liquid Crystal Society and other readers will be aware that *Liquid Crystals Today* has been published as a joint venture with Taylor & Francis Publishers since the beginning of 1995. This arrangement has allowed the ILCS to keep membership fees to a minimum, while maintaining a regular high quality news letter for its members. The initial agreement with Taylor & Francis was for a period of 5 years, which comes to an end in 1999, and it is appropriate to begin a review of *Liquid Crystals Today* and its function within the ILCS and the wider liquid crystal community.

Under the ByLaws of the ILCS, the Society does not publish original scientific research, but over the years since its inception in 1990, *Liquid Crystals Today* has published many review and feature articles of high quality, as well as news items and book reviews. Viewed as a scientific publication in the liquid crystal arena, *Liquid Crystals Today* is of course small, having a present annual size of 64 pages. However in terms of circulation and readership, *Liquid Crystals Today* is significant. Circulation is 1000–2000, and because of its modest size, most issues will be read cover to cover. If liquid crystal scientists wish to communicate their ideas to a wide audience, who will actually read their articles, then *Liquid Crystals Today* provides a highly effective vehicle. Effective and widespread communication should be the objective of all scientists, but in the new world order the scientific community is less confident, and is now obsessed by assessment. Authors will only publish material in journals that make a maximal contribution to the assessment, often through the citation index. *Liquid Crystals Today* does not have a citation rating, although perhaps it

should, but its great strength is that its articles are actually read, and many are kept for reference.

So the questions to be addressed by the Editorial Board of *Liquid Crystals Today* and the Board of Directors of the ILCS are 'Does *Liquid Crystals Today* have a future?', and if so 'What form will it take after the year 2000?' Of course these questions have to be answered both in the contexts of developments in publishing, and the future of liquid crystal science generally. The review will take place over the coming months, culminating in a discussion paper at the next ILCC in Strasbourg 1998. If the readers have any views on the future of *Liquid Crystals Today*, then they are invited to submit them to the editor.

This issue of *Liquid Crystals Today* is the last for 1997, and the Editor wishes all readers the best of liquid crystal activities for next year as the millennium approaches. Perhaps next year is the time to reorder our scientific priorities, and if you seek wide circulation and informed readership for your liquid crystal article, why not send it to *Liquid Crystals Today*. Articles are of course refereed before publication by members of the Editorial Board or other distinguished scientists.

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## Morphology Development in Liquid-Crystal/Polymer Mixtures

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**D**ispersions of a small amount of polymer in a liquid crystal matrix (polymer-stabilized liquid crystals, or PSLCs) [1–3] have shown considerable promise for liquid crystal display applications [4–6], in large part because of the polymer networks that form [7–10]. These networks have high surface areas and consequently tend to stabilize liquid crystal order efficiently, even at low concentration. There are now several experimental studies of the effects of various factors on the morphology of the networks [10–12]. For example, the networks evolve from dilute bead-like structures (see figure 1(a)) to dense, cross-linked fibrillar networks (see figure 1(b)) as a function of curing time [12]. To date, however, little is understood theoretically about the factors that control the morphology of the networks. Understanding these systems is difficult because the fabrication of liquid-crystal/polymer dispersions involves several non-equilibrium processes. These materials are typically made by photopolymerization of monomers dissolved in an ordered phase of the liquid crystal

*(continued on page 2)*

(typically nematic or cholesteric). As the polymerization proceeds, the polymer tends to phase separate from the liquid crystal, forming an isotropic phase rich in polymers coexisting with an ordered phase rich in liquid crystals. In true equilibrium, these two phases would separate on a macroscopic scale. Long before the system reaches that point, however, the polymerization process freezes in a cross-linked network of polymer-rich domains. A theoretical description of this process must therefore contain the kinetics of polymerization,

phase separation and phase ordering. This is a formidable task, because the three processes are interconnected: for example, the polymerization rate depends on the local concentration, which depends on the kinetics of phase separation [13–15]. Here I will focus only on the interplay of the kinetics of phase separation and phase ordering.

The first question to ask when considering the interplay between phase separation and ordering is: when does ordering really matter? There are cases where orientational order has very little

effect on domain morphology. For example, it is clear that ordering kinetics plays almost no role when the starting state and ending state are both isotropic. Most polymer dispersed liquid crystals (PDLCs), which contain a majority of polymer, and relatively little (typically around 30%) liquid crystal, are systems that fall into this category. However, even if one of the phases at the end of the phase separation process is ordered, the morphology may still be insensitive to ordering. The relevant question is whether the system is initially unstable to

(continued on page 3)

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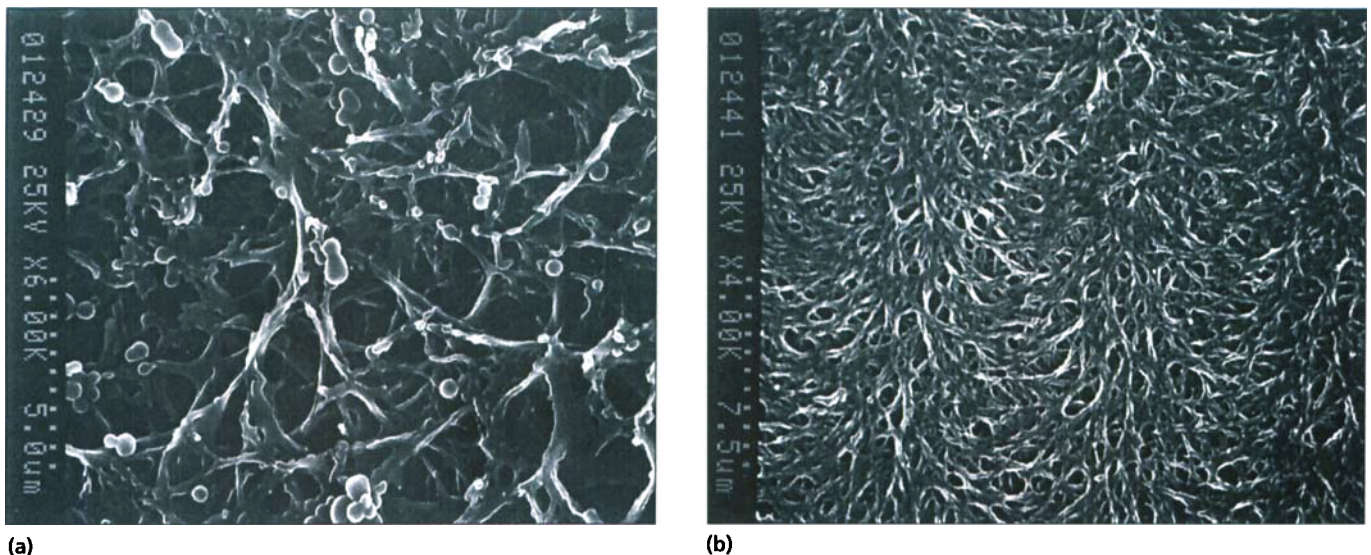
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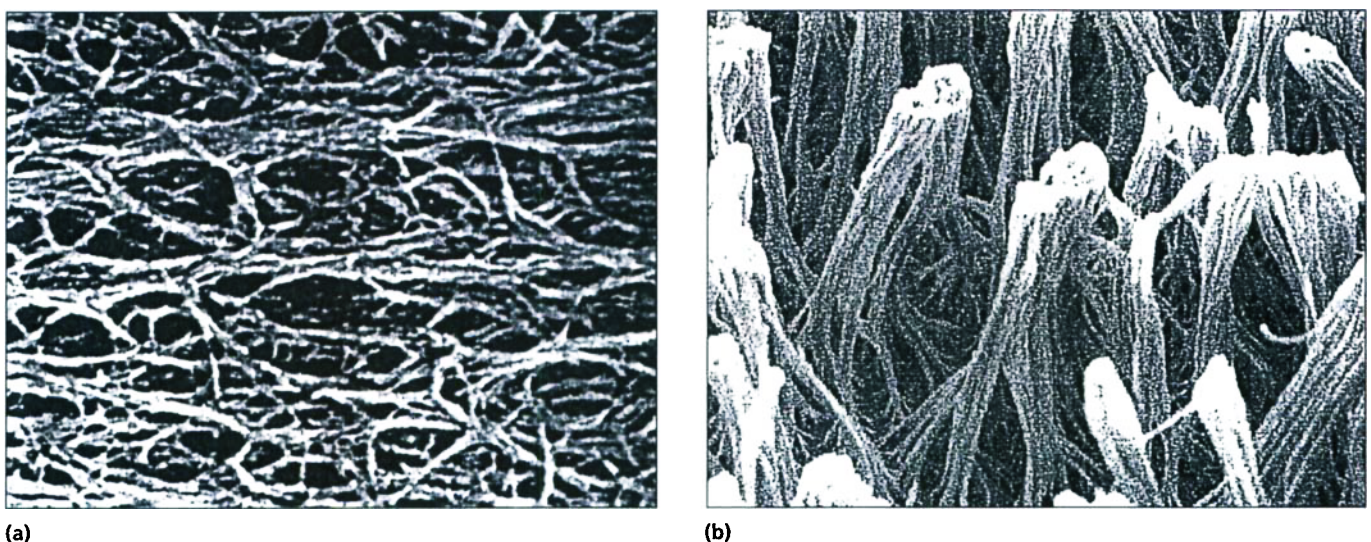
**(a)** **(b)**  
 Figure 1. Effect of UV curing time on the morphology of polymer networks in PSLCs. These are SEM images of polymer networks that have been cured in the cholesteric phase for (a) 5 min and (b) 30 min, respectively. At the shorter curing time (a), a network composed of strands and beads has formed. After 30 min of polymerization (b), the network is much denser and beads are no longer visible. Courtesy of I. Dierking, L. L. Kosbar, A. C. Lowe and G. A. Held, [12].

phase ordering. As a rough rule of thumb, ordering kinetics significantly affects domain morphology when the starting state is ordered, or when one of the final phases is ordered and the starting state is unstable to phase ordering as well as phase separation. In the case of polymer-stabilized systems, where the starting state is nematic or cholesteric, orientational ordering can play an especially significant role in morphology development. Experiments

show that the polymer networks that develop in PSLCs at low polymer concentrations can be modelled as fibres or bundles of fibres that run parallel to the nematic director [8–12] (see figure 2). At higher polymer concentrations, experiments suggest that the morphology may be sheet-like [16]. Thus, it appears that ordering gives rise to anisotropic domain structures.

There are several factors, both thermodynamic and kinetic, that can

influence domain shape anisotropy. For example, the interfacial tension between an isotropic and nematic phase is anisotropic, so domain shapes tend to distort to lower the surface free energy. This thermodynamic effect is largest for domains that are comparable to the length  $k/\gamma$ , where  $\gamma$  is the interfacial tension between the two final phases and  $k$  is the elastic constant. This length scale is often comparable to the characteristic length scale at which



**(a)** **(b)**  
 Figure 2. Network morphology for PSLCs cured in the nematic phase. These are SEM images of polymer networks formed in (a) the homogeneous nematic starting state, and (b) nematic homeotropic state under an applied electric field, with the normal of the substrate tilted a few degrees away from the incident electron beam. These images show that the fibrils tend to run parallel to the local nematic director. Courtesy of Y. K. Fung, D. K. Yang, S. Ying, L. C. Chien, S. Zumer and J. W. Doane [8].

spinodal decomposition begins. During the later stages of the phase separation process, however, the average domain size grows with time. Once it becomes large compared to  $k/\gamma$ , the domains should become more isotropic in shape. This is because the interfacial energy dominates the bulk elastic energy in large domains. In order to obtain a highly fibrillar morphology, it is therefore important to arrest the phase separation process (e.g. by forming a cross-linked network) before the domains grow too large.

At early times following the onset of spinodal decomposition, the kinetics can *amplify* the thermodynamic tendency to form anisotropic domains. The growth rate for the concentration difference between the two emerging phases depends exponentially on the thermodynamic driving force. This driving force is anisotropic. For example, consider the case where the liquid crystals prefer parallel anchoring at the polymer interface. Suppose we impose a composition perturbation of the form  $\sin(\mathbf{q}\cdot\mathbf{r})$  around the starting, uniform state. This perturbation will grow most rapidly if  $\mathbf{q}$  is oriented perpendicular to the director  $\mathbf{n}$ , because the free energy is lower if the liquid crystals orient perpendicular to the composition gradients (i.e. parallel to the developing interfaces). In other words, the composition difference between the developing isotropic, polymer-rich regions and nematic, liquid-crystal rich regions will increase most rapidly for  $\mathbf{q}$  perpendicular to the director. This may lead to fibrils or sheets oriented parallel to the director. Another important effect is the anisotropy in the diffusion coefficient: the liquid crystal molecules diffuse more rapidly in the direction of the director. This also tends to favour anisotropic domains parallel to the director.

Our aim is to capture these effects and study the morphological development theoretically. We have constructed a framework for studying the kinetics of phase separation into an isotropic phase rich in polymer coils and a nematic phase rich in rods [20]. In this case, both composition and orientational density evolve in a coupled fashion as functions of position and time. The order parameters can change with time because of diffusive motion. Thus, the composition can change due to translational diffusion of rods and coils, and the orientational density can change due to translational diffusion of rods and coils, or to rotational diffusion of rods. The standard Cahn–Hilliard approach to study phase separation kinetics alone is to construct an equation of motion for the composition order parameter of the form [17]:

$$\text{Model 1: } \frac{\partial\phi(\mathbf{r},t)}{\partial t} = \Gamma_{\phi\phi} \nabla^2 \frac{\delta F[\phi(\mathbf{r},t)]}{\delta\phi} \quad (1)$$

Here, the composition order parameter  $\phi(\mathbf{r},t)$  is the local volume fraction of one of the species,  $\Gamma_{\phi\phi}$  is a mobility coefficient that depends on the diffusion coefficients of the two species in the mixture, and  $F[\phi]$  is a free energy functional that drives the system towards equilibrium. The natural generalization to include the possibility of nematic ordering is to introduce two coupled equations of motion for  $\phi(\mathbf{r},t)$  and the orientational density  $S(\mathbf{r},t)$ :

$$\frac{\partial\phi(\mathbf{r},t)}{\partial t} = \Gamma_{\phi\phi} \nabla^2 \frac{\delta F[\phi,S]}{\delta\phi} \quad (2)$$

Model 2:

$$\frac{\partial S(\mathbf{r},t)}{\partial t} = -\Gamma_{SS} \frac{\delta F[\phi,S]}{\delta S}$$

This approach has been adopted by Dorgan [18] and by Lansac *et al.* [19]. Our approach is similar to theirs and differs mainly in that we have taken into account the full tensorial nature of the orientational density,  $\mathbf{S}(\mathbf{r}, t)$  [20]. The resulting equations have the form

$$\frac{\partial\phi(\mathbf{r},t)}{\partial t} = \Gamma_{\phi\phi} \nabla^2 \frac{\delta F[\phi,\mathbf{S}]}{\delta\phi} + \Gamma_{\phi S}^{ij} \frac{\delta F[\phi,\mathbf{S}]}{\delta S^{ij}}$$

Model 3:

$$\frac{\partial S^{ij}(\mathbf{r},t)}{\partial t} = \Gamma_{\phi S}^{ij} \frac{\delta F[\phi,S]}{\delta\phi} - \Gamma_{SS}^{ijkl} \frac{\delta F[\phi,S]}{\delta S^{kl}}$$

(3)

Model 2 is applicable only when the nematic director is uniform throughout the sample. For example, it describes domain growth in thin samples confined between parallel plates that enforce a uniform director field, or in bulk samples under strong electric fields. The advantage of Model 3 is that it applies to the more general case, where the nematic director can vary in space. This is *crucial* in order to capture anisotropic domain growth in bulk samples. In addition, we have used dynamical mean field theory to *derive* Model 3, and therefore are able to express all of the mobility coefficients ( $\Gamma_{\phi\phi}$ ,  $\Gamma_{\phi S}^{ij}$ ,  $\Gamma_{SS}^{ijkl}$ ) in terms of single-macromolecule quantities, such as the translational diffusion coefficient of the coil, the rotational diffusion coefficient of the rod, and the parallel and perpendicular translational diffusion coefficients of the rod [20]. Note that we also find extra cross terms (proportional to  $\Gamma_{\phi S}^{ij}$ ) in the equations of motion. The mobility coefficients  $\Gamma_{\phi S}^{ij}$  are non-local and arise from the extended shapes of the rods and coils. Similar non-local terms appear in equations derived by Shimada *et al.* for the case of rod/solvent mixtures [21].

The free energy functional  $F[\phi, \mathbf{S}]$  in Model 3 controls the phase separation kinetics. It has been calculated within mean-field theory from microscopic models of rods and coils in terms of single-macromolecule quantities such as the molecular weights [22]. By varying the molecular weights of the rods and coils, Model 3 can be applied to a variety of systems, ranging from small-molecule liquid crystal/flexible polymer mixtures to liquid crystalline polymer/solvent mixtures. The model depends on only two phenomenological parameters, characterizing the strengths of the isotropic and anisotropic interactions, respectively; fortunately, these two parameters can be estimated from experimental phase diagrams [23]. The free energy can be written as the sum of three terms. The first term is the Flory–Huggins free energy, which depends only on the volume fraction  $\phi(\mathbf{r}, t)$ , and describes phase separation between two polymeric components (in this case, a rodlike species and a flexible polymer coil species). The second term is the Landau–de Gennes free energy, which describes the isotropic/nematic transition in terms of the orientational order parameter  $S(\mathbf{r}, t)$ . In this case, the coefficients of the Landau expansion are calculated functions of the rod length and  $\phi(\mathbf{r}, t)$ ; the dependence on  $\phi(\mathbf{r}, t)$  couples the two order parameters and leads to complex phase



diagrams [23]. Finally, the third term in the free energy describes the costs of gradients in  $\phi(\mathbf{r}, t)$  and  $S(\mathbf{r}, t)$ . The gradients have the form

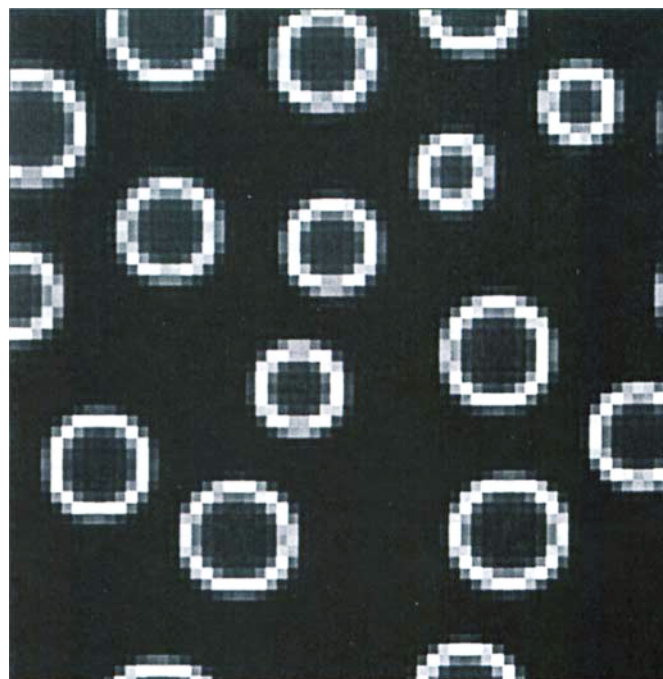
$$F_{grad} = \frac{1}{2} M(\nabla\phi)^2 - L_0\partial_i\phi\partial_j S^{ij} + \frac{1}{2} L_1(\partial_k S^{ij})^2 + \frac{1}{2} L_2\partial_i S^{jk}\partial_j S^{ik} \quad (4)$$

where the coefficients  $M, L_n$  are also calculated functions of the rod length and  $\phi(\mathbf{r}, t)$ . The first term in equation (4) controls the cost of spatial gradients in  $\phi(\mathbf{r}, t)$ . The last two terms determine the cost of gradients in the orientational density, and are related to the Frank constants. Note that the magnitude of  $M$  determines the width of the interface preferred by the concentration profile; the larger the value of  $M$ , the sharper the interface. Similarly,  $L_1$  and  $L_2$  determine the width of the interface preferred by the orientational density. These preferred widths can be quite different, depending on the contour lengths of the rods and coils, and the interaction parameters. The final interfacial width is determined by a compromise between the coupled order parameters. Finally, the coefficient  $L_0$  couples gradients in concentration to gradients in orientational density. This term is responsible for the anchoring conditions at the interface; rods prefer not to penetrate through the interface into the coil-rich phase, so they tend to run along the interface, giving rise to parallel orientation at the interface. This effect has been observed even for flexible polymers at interfaces separating two isotropic phases [24], and is stronger at isotropic/nematic interfaces [25, 26].

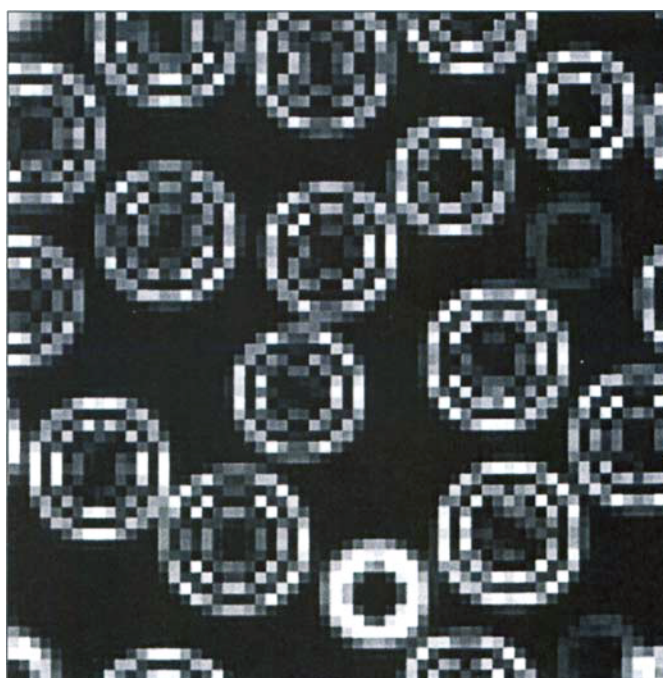
Models 2 and 3 have been solved analytically at early times following a quench into the two-phase region, when the composition difference between the two emerging phases is still small [18–20]. The early-time analyses show that there is indeed a strong coupling between composition and orientation that can lead to faster growth in the direction of the local nematic director. At later times, the equations must be treated in their full nonlinear form, and can only be solved numerically. Lansac *et al.* have carried out numerical solutions of Model 2 for a two-dimensional system at late times, for the case where the nematic director is uniform and in the plane of study [19]. Their calculation would apply to thin samples sandwiched between two plates with parallel anchoring surfaces. They do indeed find anisotropic domains (see figures 13 and 14 in [19]); in their case, the long-axis of the domains is always parallel to the uniform nematic director.

At this point, it is still unclear whether Model 3 can lead to the dramatically fibrillar morphologies observed experimentally. Preliminary numerical results show that ordering can lead to noticeable changes in domain morphology [27]. Figure 3 shows the morphology at two different times following a quench from the isotropic mixed state [27]. At an intermediate time, the domains rich in liquid crystal are circular, with parallel ordering at the interface and a defect at the centre (see figures 3(a) and (b)). The degree of ordering inside is extremely weak because the concentration of the liquid crystal is not yet high enough. At later times, however, the defects are expelled from the domain, leaving strong, uniform ordering within elongated domains, and very weak ordering at the two extreme ends (see figures 3(c) and (d)). Note that different elongated domains are oriented in

different directions; this is possible in Model 3 because we have used the tensorial form of  $S(\mathbf{r}, t)$ . The structure is still far from being fibrillar as in figure 2, however. If we quench from an ordered mixed state, we do find anisotropic structures at intermediate times, with fibrils running parallel to the director



(a)

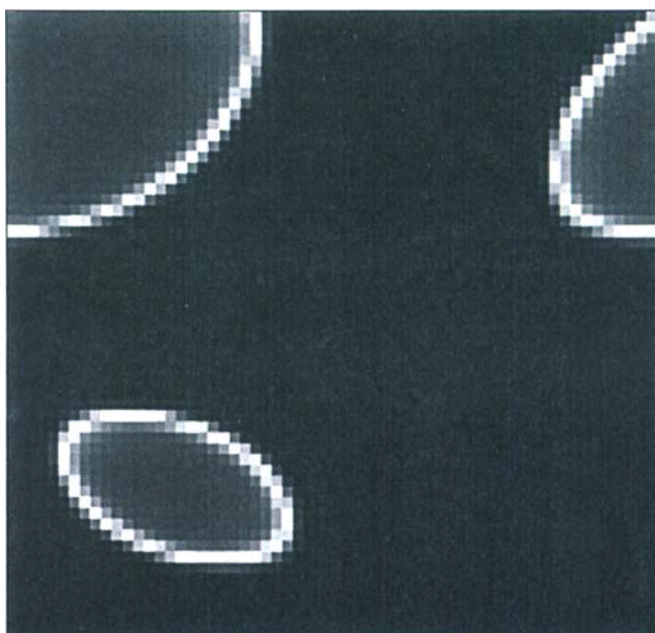


(b)

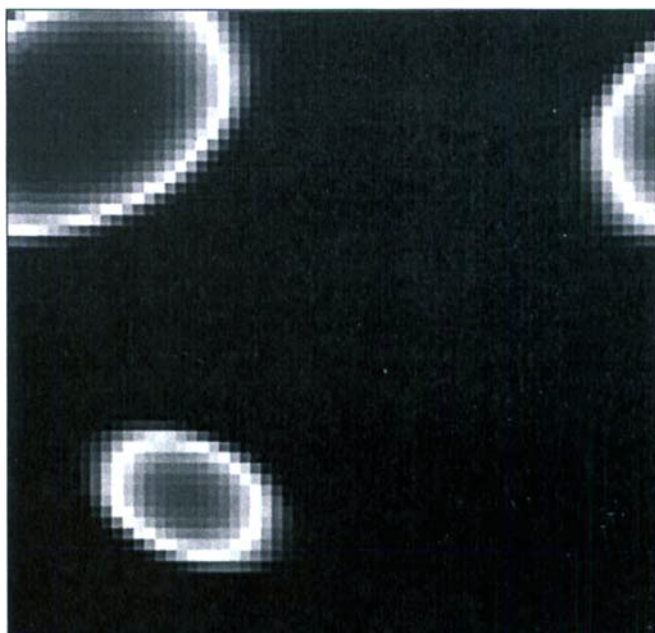
Figure 3. Numerical solutions of Model 3 in two dimensions for a quench from an isotropic starting state.

(a) The concentration  $\phi(\mathbf{r})$  of liquid crystal molecules at an intermediate time following the quench. The grey scale ranges from dark to light for  $0.068 \leq \phi \leq 0.677$ .

(b) The magnitude of the orientational density  $S(\mathbf{r})$  at the same intermediate time; the grey scale ranges from dark to light for  $0 \leq S \leq 0.008$ .



(c)



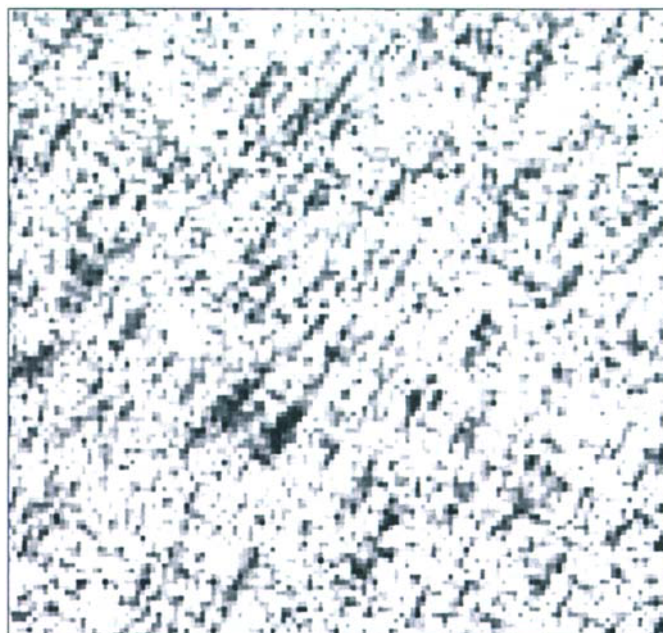
(d)

*Figure 3 continued*

(c) The concentration  $\phi(\mathbf{r})$  at a later time; the grey scale corresponds to  $0.058 \leq \phi \leq 0.776$ .

(d) The orientational density  $S(\mathbf{r})$  at the same later time; the grey scale corresponds to  $0 \leq S \leq 0.02$ . At intermediate times, ordering does not affect domain morphology. At later times, ordering causes domains to elongate in the direction of nematic order.

(see figure 4) but the fibrils tend to break up into elongated droplets once the composition difference between the two phases becomes appreciable [27]. Perhaps it is necessary to include polymerization kinetics in order to reproduce the fibrils that are observed experimentally, but much more numerical work remains to be done on Model 3 to explore all the factors that control morphology.



**Figure 4.** Numerical solution of Model 3 for a quench from an ordered starting state. The nematic director in the starting state is imposed at an angle of  $3\pi/10$ . The resulting domains at an intermediate time after the quench are fibrillar, oriented along the director. However, the composition difference between the two phases is still quite small at this stage of the phase separation process; the grey scale ranges from  $0.589 \leq \phi \leq 0.611$ .

The development of polymer/liquid-crystal dispersions for applications has raised many fundamental questions that pose conceptual and technical challenges to theorists. Even when polymerization kinetics are neglected, the interplay of ordering kinetics and phase separation kinetics already leads to rich phenomena that are not yet fully understood. The hope is that fundamental understanding of the factors that control morphology development, while interesting in its own right, will also lead to enhanced control over the microstructure of these dispersions.

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**Editor's note:** The illustrations to this article were submitted in colour. Regrettably it has not been possible to reproduce them in colour, but colour copies can be obtained from the author.

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