Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Golovataya, N. M. , Kurik, M. V. and Lavrentovich, O. D.(1990) 'Self-organization of polymer dispersed nematic droplets', Liquid Crystals, 7: 2, 287 — 291
To link to this Article: DOI: 10.1080/02678299008029216
URL: http://dx.doi.org/10.1080/02678299008029216

LIQUID CRYSTALS, 1990, VOL. 7, NO. 2, 287–291

Self-organization of polymer dispersed nematic droplets

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(*Received 5 October 1989; accepted 14 November 1989*)

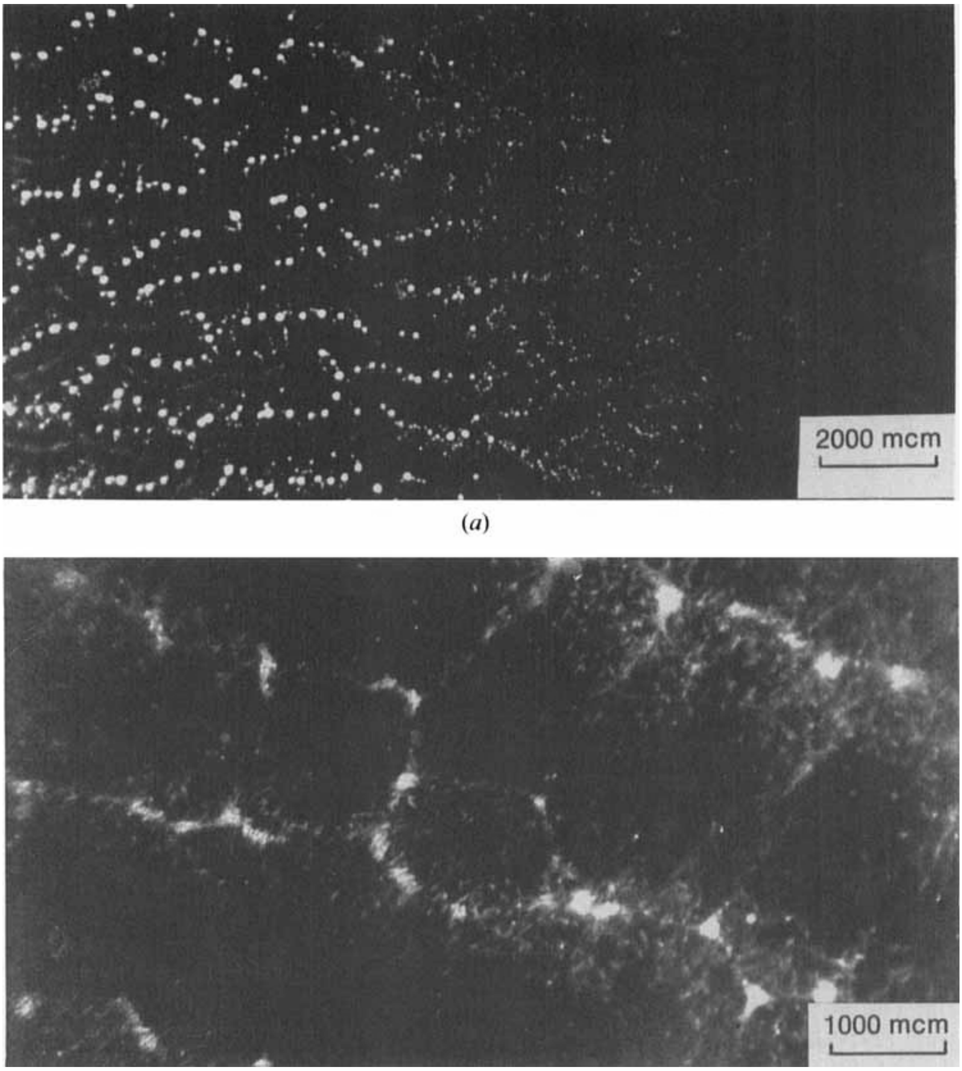
Self-organization of polymer dispersed nematic droplets has been observed experimentally. Self-organization is manifested in the spatial periodic distribution of drop clusters as well as in the orientational ordering of the symmetry axes of the droplet structures. The possible elastic and electrostatic mechanisms for the phenomena are discussed.

Dispersions of liquid-crystalline droplets are of interest because of their unusual physical properties [1] and practical applications [2]. In this preliminary communication we report experimental observations of a new effect of droplet self-organization in polymer dispersed liquid crystal films. Self-organization is manifested, firstly, in the spatial periodic distribution of droplet clusters and secondly, in the orientational ordering of the symmetry axes of the drop structures.

The samples were prepared in the usual manner [3]. Liquid crystal (4-*n*-pentyl-4′-cyanobiphenyl (5CB)) was dissolved with the polymer (a polyamid resin) in a common solvent (acetone or ethanol). The weight ratio was 1 : 1 : 8 in mixture I and 1·4 : 1 : 8 in mixture II. The resulting isotropic solution was coated on a transparent glass substrate, forming a liquid film 1–2 mm thick. The upper boundary of the film was left free and the solvent was evaporated in a clean environment. The textures of the system were investigated with a polarizing microscope in transmitted light.

Evaporation of the solvent results in convective flows of liquid and the convective patterns are similar to those observed in evaporated pools of isotropic liquids, see, for example, [4]. Evaporation of the solvent moves the system towards the miscibility gap and this results in nematic droplet formation. The droplets are involved in the convective flows. There are two types of trajectories: rolls and cells (see figure 1). As a rule, rolls were observed in mixture I and cells were observed in mixture II. When the solvent has evaporated, the flow slows down and motionless patterns with rolls or cells form (see figures 2–4).

Distributions of the drops possess two types of ordering. The first consists of a spatial periodic distribution of drop clusters (see figure 2), the second consists of orientational ordering and arises in the roll structures (see figures 3 and 4). Orientational order is manifested by the ordered inner structure of the nematic drops. These symmetric structures occur in practically all of the drops. The axes **N** of the drop structures are oriented along the common direction **M**. In some cases **M** does not coincide with the roll direction **R**; for the structures shown in figure 3, **M** || **R**, but for the structures in figure 4, **M** and **R** make an angle of 30–60°. We discuss briefly the



(*a*)

(*b*)

Figure 1. Formation of nematic drops during solvent evaporation: (*a*) roll clusters in mixture I; the concentration of the solvent increases from the left to the right hand side of the sample; (*b*) cell clusters in mixture II.

possible mechanisms for the self-organization observed in the polymer-dispersed liquid crystal films.

The spatial ordering of the drops is a direct consequence of the convective motion in the system during the early stages of polymer film preparation. The convective patterns themselves arise due to two mechanisms: first the surface tension driven convection (the Marangoni effect) and secondly the buoyancy driven convection. Both of these mechanisms depend on the fact that solvent evaporation cools the liquid surface thereby increasing the surface tension as well as the density and leading in each instance to a potentially unstable stratification of the mixture. In general, these mechanisms in polymer dispersed liquid crystal films are analogous to those in the

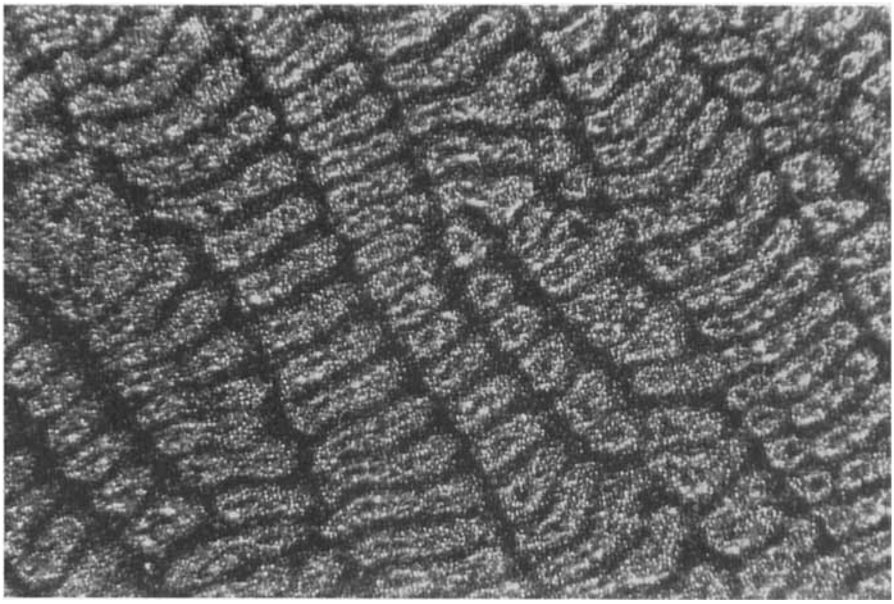


Figure 2. Cell clusters in a polymer dispersed liquid crystal film, mixture II.

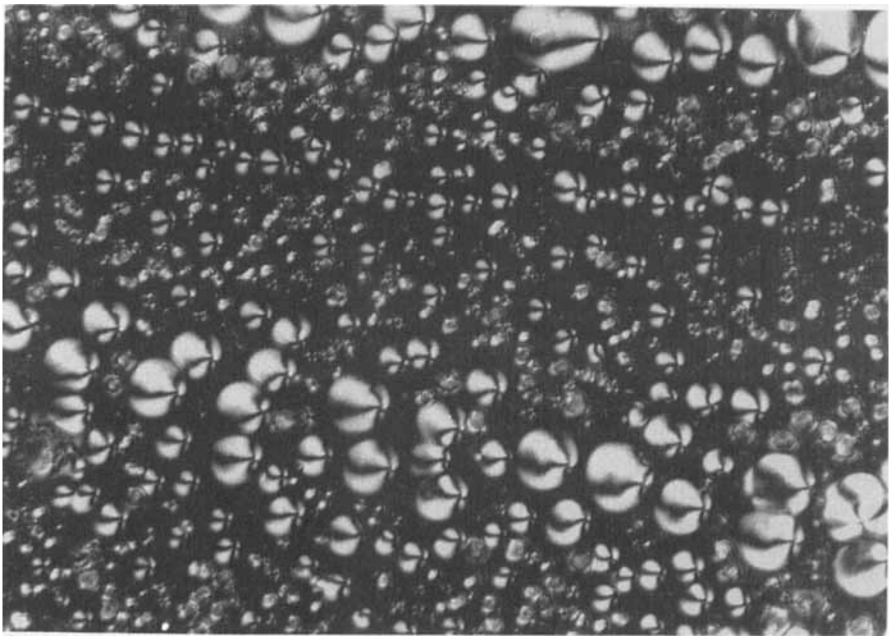


Figure 3. Roll clusters in a polymer dispersed liquid crystal film, mixture I; the roll direction coincides with the direction of the droplet orientation.

pools of evaporating pure liquids or binary solutions from which one component is desorbed, see, for example, [4].

Alternatively, the nature of the orientational order should be closely connected with the properties of the liquid-crystalline component, in particular, with the

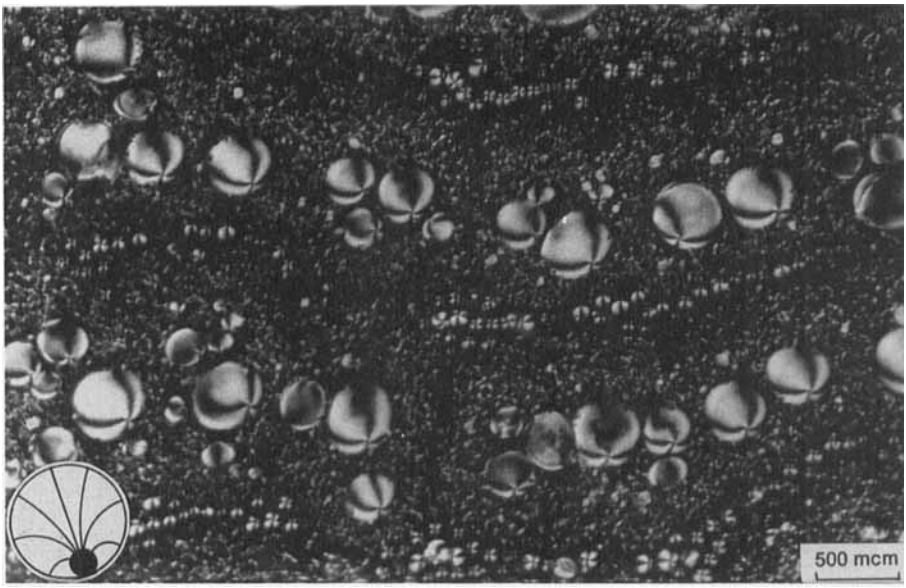


Figure 4. Roll clusters in a polymer-dispersed liquid crystal film, mixture II; the roll direction does not coincide with direction of the droplet orientation.

non-uniform distribution of the director **n** inside the drop. There are at least two possible causes for the orientational order in the system of the nematic drops: elastic deformations of the drop shapes and electrostatic interaction between the drops. The former is connected with possible deformations of the drop cavities due to deformation of the polymer binder. In deformed drops the axis **N** will prefer to orient along the axis of the drop cavity, where the elastic energy is minimized [5, 6]. The elastic energy U_{el} is expected [5] to be $\sim qKR$, where K is the effective elastic constant, $q = (a^2 - b^2)/(a^2 + b^2) < 1$, a and b are the long and short axes of the cavity, respectively and R is the average radius of the drop. Taking $K = 10^{-11}$ N and $R = 100\,\mu$m, we obtain $U_{el}/kT \sim 1$ even for negligibly small $q = 4 \times 10^{-5}$. Thus in real systems the orientation of **N** may be non-degenerate because of deformation of the cavities.

The convective motions may be responsible for the elongation of the drop cavities in one direction **M**, and thus, for the orientational ordering of the drop axes along **M**. However, this possibility cannot explain directly the observation of different values of the angle between **M** and the roll direction **R** (see figures 3 and 4). Moreover, in a number of drops the axis **N** does not coincide with the observed axis of the drop cavity (see figures 3 and 4).

Electrostatic interaction of the drops may be caused by van der Waals forces [7] or (and) by the flexoelectric effect [8, 9]. The energy U_{dd} of the van der Waals interaction between two anisotropic drops should depend on the angle between the axes of the drops. The magnitude of U_{dd} would be expected [7] to be comparable to kT for drops of $R = 100\,\mu$m and at a distance of $10\,\mu$m. Unfortunately, in our experiment the distances between some drops are of several tens of a micrometer. Morever, there is another difficulty namely the polar character of the drop ordering (**M** is a vector rather than the director). Polar ordering as well as the long range of the interaction may be explained by the flexoelectric effect. Drops with a geometry similar to that depicted in figures 3 and 4 possess a dipole moment [9] $\mu = \lambda(e_1 - e_3)Rh$, where e_1 and e_3 are the flexoelectric constants and h is the height of the drop. The corresponding coupling constant is $\lambda = \mu^2/4\pi\varepsilon_0 d^3 kT$, where d is the separation. For room temperatures, $e_1 = -e_3 = 10^{-11}$ C/m, $Rh = 10^3\,\mu$m and for $d < 1000\,\mu$m we find $\lambda > 1$. As a result, for sufficiently pure substances [9], the flexoelectric effect may explain the observed phenomena of the self-organization of nematic drops, especially the polar character of the ordering. However, the phenomena observed deserve a detailed study.

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