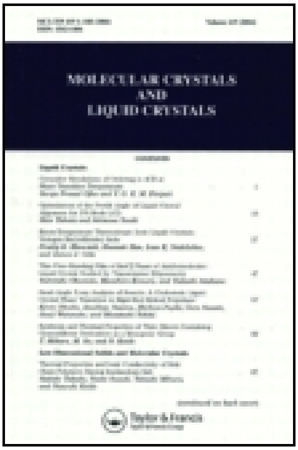

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006

To cite this article: Masayoshi Todorokihara, Koji Seike, Hiroyoshi Naito & Ou-Yang Zhong-Can (2001): Pattern Formation of Two-Dimensional Smectic-A Filaments, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 364:1, 403-410

To link to this article: http://dx.doi.org/10.1080/10587250108025009

Mol. Cryst. and Liq. Cryst., 2001, Vol. 364, pp. 403-410
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Pattern Formation of Two-Dimensional Smectic-A Filaments

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Electric field-induced pattern formation of two-dimensional smectic-A filaments have been observed in the binary mixture of octyloxycyanobiphenyl and dodecyl alcohol. As the mixture is cooled below its transition temperature, the smectic-A phase grows into a collection of filaments directly from an isotropic phase. Without the electric-field application, these filaments, which have fixed radius, buckle three-dimensionally as they grow. When the liquid crystal samples are subjected to electric field, the three-dimensional patterns of smectic-A filaments are changed to two-dimensional periodic wavy patterns of the filaments. The two-dimensional pattern is formed on a plane perpendicular to electric field. To understand the electric field-induced two-dimensional pattern formation process, the shape equations for the filaments have been derived by a variation of the sum of Gibbs free energy, curvature elastic energy, surface energy, and dielectric energy. An analytical solution to the shape equations are derived, and shown to be in good agreement with the experimental observation.

Keywords: pattern formation; smectic-A filaments; plane curves; electric field; shape equations

INTRODUCTION

Thermo-temporal evolution of smectic-A phase is a geometrically interesting example of self-organization processes. A smectic-A phase grown from an isotropic phase first appears a collection of filaments. These filaments have a fixed radius and buckle continuously as they grow. The length of these filaments is seen to grow exponentially, which suggests that the filament grows locally at a constant rate [1]. Such a growth process is very different from the one usually seen in similar situations, such as the growth of the nematic phase during the isotropic-nematic transition and the growth of ice crystals during the liquid-solid transition. Additionally the filaments exhibit spectacular dynamics, and hence a study of smectic-A filaments has attracted much attention [2-7]. We have studied such pattern formation of a smectic-A phase in an isotropic phase both experimentally and theoretically, and have found that the patterns of smectic-A phase can be explained in terms of a variational calculation of a sum of Gibbs free energy, curvature elastic energy and surface energy [3]. It is expected that the electric-field application causes dramatic changes in patterns of a smectic-A phase in an isotropic phase because of large dielectric anisotropy of liquid crystals. However, the effects of electric field on the pattern formation of a smectic-A phase in an isotropic phase have not yet been fully investigated [4].

In this paper, we report the pattern formation of smectic-A filaments under electric-field application. We find that the pattern formation processes of smectic-A filaments are greatly influenced by electric field. The three-dimensional growth patterns are changed into the two-dimensional periodic patterns, characterized by plane-curves. It is shown that the pattern formation of the two-dimensional periodic filaments under the electric-field application can be explained within the theoretical framework that we have proposed earlier [3,5].

EXPERIMENT

The liquid crystal (LC) material used here was octyloxycyanobiphenyl (8OCB). Dodecyl alcohol (DODA) was mixed with 8OCB (a molar ratio of 8OCB to DODA was 4 to 6) to suppress the nematic phase and to observe a smectic-A phase in an isotropic phase. Figure 1 shows the phase diagram of the mixture of 8OCB and DODA. There appears a coexisting region of the smectic-A and the isotropic phases. The LC cells of dimensions 10 mm$\times$10 mm and of thickness 30-80 μm bound by indium-tin oxide (ITO) precoated glass plates were prepared. The ac square-wave electric field (2 kHz) was applied to the cells. This frequency is sufficient to overcome the effects of ionic impurities without essentially affecting the value of the dielectric anisotropy of 8OCB ($\Delta\varepsilon$). The sample temperature

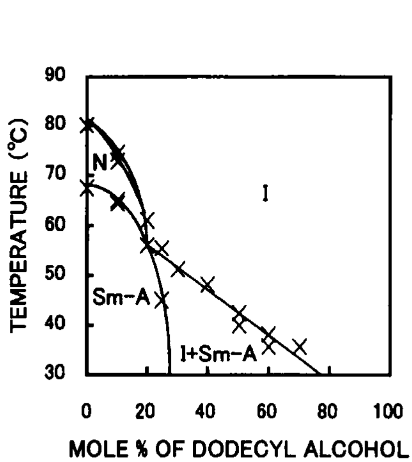


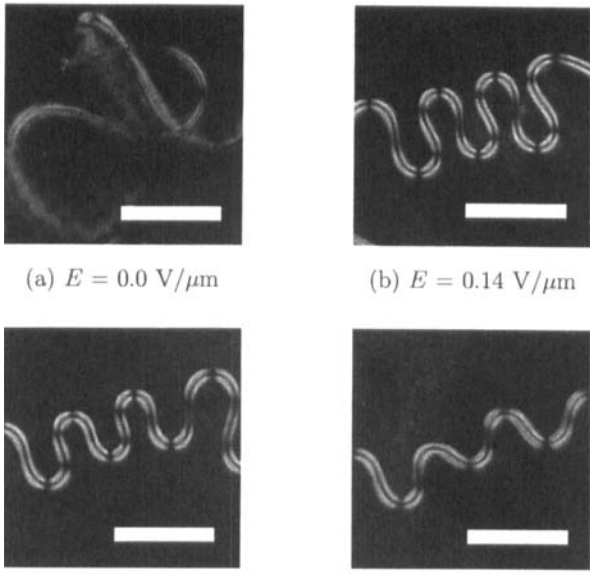
FIGURE 1 Phase diagram for the mixture of 8OCB and DODA [6].

was controlled using a hot stage (Instec HS1-i). The pattern formation of the smectic-A phase was observed with a polarizing microscope (Nikon X2TP-11) equipped with a digital camera (Olympus DP-11).

RESULTS

As the mixture is cooled from the isotropic phase at a cooling rate of 0.1 °C/min, the Sm-A phase initially appears at ~40 °C in the form of a number of spherical droplets which grow in size and then start elongating into straight filaments. On cooling further, filaments buckle continuously and become the three-dimensional entangled patterns (Fig. 2(a)). Computer simulation of the polarizing microscope images of smectic-A filaments showed that a smectic-A filament can be described by a tube with the outer and inner surfaces.

To study the influence of electric field on the pattern formation of the smectic-A filaments, the ac electric field was applied in the direction perpendicular to the ITO glass surfaces. When the LC cell is subjected to electric field of 0.14 V/μm, a drastic change in patterns of smectic-A filaments is observed; the three-dimensional pattern of smectic-A filaments is changed to the two dimensional pattern of smectic-A filaments, which is



(a) $E = 0.0$ V/μm (b) $E = 0.14$ V/μm

(c) $E = 0.15$ V/μm (d) $E = 0.16$ V/μm

FIGURE 2 Patterns of the smectic-A filament observed with the analyzer and polarizer crossed (a) $E = 0.0$ V/μm, (b) $E = 0.14$ V/μm, (c) $E = 0.15$ V/μm, and (d) $E = 0.16$ V/μm. The bars indicate 40 μm.

formed on a plane perpendicular to electric field. We can see in Fig. 2(b) that the smectic-A filaments are in the form of almost periodically buckled filaments and on a plane perpendicular to the electric field. The periodically buckled filaments are equilibrium shapes in the sense that the shapes of the filaments are essentially unchanged at a constant temperature and an electric field. We also see in Fig. 2 that the amplitude of the periodically buckled filaments is decreased and the periodicity along the growth direction is increased with increasing electric field.

DISCUSSION

In order to discuss the equilibrium shapes of smectic-A filaments subjected to electric field, we derive the shape equations by the variation of the free energy of a Sm-A filament in an isotropic phase. The free energy of a smectic-A filament (F) may be expressed as the sum of the following four terms: (1) the volume free energy change due to isotropic–smectic-A transition (F_V)

$$F_V = -g_0 V = -g_0 \int \mathrm{d}V = -\pi g_0(\rho_o^2 - \rho_i^2) \int_C \mathrm{d}s, \tag{1}$$

where g_0 is the difference in the Gibbs free-energy densities between the isotropic and the smectic-A phases, V is the volume of the smectic-A filament, and ρ_o and ρ_i are, respectively, the outer and inner radii of Sm-A filament; (2) the surface energy of the smectic-A/isotropic interface (F_A)

$$F_A = \gamma A = \gamma \oint \mathrm{d}A = 2\pi(\rho_o + \rho_i)\gamma \int_C \mathrm{d}s, \tag{2}$$

where γ is the isotropic–Sm-A interfacial tension, A is the interface area between the isotropic and the smectic-A phases; (3) the curvature elastic energy of a smectic-A filament (F_C)

$$F_C = k_c \int_C \kappa(s)^2 \mathrm{d}s + \pi k_{11} \ln\left(\frac{\rho_o}{\rho_i}\right) \int_C \mathrm{d}s, \tag{3}$$

where k_{11} is the splay elastic constant, $k_c = \pi k_{11}(\rho_o^2 - \rho_i^2)/4$, $k_5 = 2k_{13} - k_{22} - k_{24}$ (k_{ij} the Oseen-Frank elastic constants), $\kappa(s)$ is the curvature defined along $\boldsymbol{r}(s)$, $\boldsymbol{r}(s)$ is the axial curve of the filament; (4) the dielectric energy of the smectic-A filament (F_D)

$$F_D = -\frac{\varepsilon_0 \Delta\varepsilon}{2} \frac{\rho_o^2 - \rho_i^2}{2} \pi \int_C \left\{E^2 - (\boldsymbol{E} \cdot \boldsymbol{t})^2\right\} \mathrm{d}s, \tag{4}$$

where ε_0 is the permittivity of free space, $\boldsymbol{E}$ is the applied electric field, $E = |\boldsymbol{E}|$, $\boldsymbol{t} = \dot{\boldsymbol{r}}(s) = \mathrm{d}\boldsymbol{r}/\mathrm{d}s$. Thus, we obtain the total energy of the smectic-A filament

$$\begin{aligned} F &= F_V + F_A + F_C + F_D \\ &= \int_C \left[k_c\kappa(s)^2 + \Upsilon(\boldsymbol{E}\cdot\boldsymbol{t})^2 + (\lambda + \Pi - \Upsilon E^2)\right] \mathrm{d}s, \end{aligned} \tag{5}$$

where $\Upsilon = \pi\varepsilon_0\Delta\varepsilon(\rho_o^2 - \rho_i^2)/4$, $\lambda = 2\pi(\rho_o + \rho_i)\gamma + \pi k_{11}\ln(\rho_o/\rho_i)$, and $\Pi = -\pi g_0(\rho_o^2 - \rho_i^2)$. The shape equations are given by the first variation of the total energy, $\delta F = 0$, with respect to the normal and the binormal direction of smectic-A filaments. After some calculation, we have the two shape equations for the Sm-A filament,

$$\begin{aligned} k_c(\kappa^3 - 2\kappa\tau^2 + 2\ddot{\kappa}) - (\lambda + \Pi - \Upsilon E^2)\kappa \\ +\Upsilon(\boldsymbol{E}\cdot\boldsymbol{t})^2\kappa - 2\Upsilon(\boldsymbol{E}\cdot\boldsymbol{m})^2\kappa = 0, \end{aligned} \tag{6}$$

$$2\dot{\kappa}\tau + \kappa\dot{\tau} + 2\Upsilon(\boldsymbol{E}\cdot\boldsymbol{m})(\boldsymbol{E}\cdot\boldsymbol{b})\kappa = 0, \tag{7}$$

where $\tau = \tau(s)$ is the torsion defined along $\boldsymbol{r}(s)$, $\dot{\kappa} = \mathrm{d}\kappa/\mathrm{d}s$, $\ddot{\kappa} = \mathrm{d}^2\kappa/\mathrm{d}s^2$, $\dot{\tau} = \mathrm{d}\tau/\mathrm{d}s$, and $\boldsymbol{m} = \boldsymbol{m}(s)$, $\boldsymbol{t} = \boldsymbol{t}(s)$ and $\boldsymbol{b} = \boldsymbol{b}(s)$ are, respectively, the normal, tangential and binormal vectors of $\boldsymbol{r}(s)$.

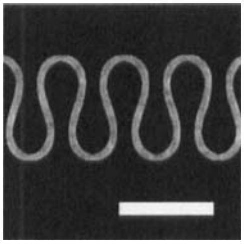
Substituting the experimental condition ($|E|$ = *constant*) and experimental results ($\boldsymbol{E}$ // $\boldsymbol{b}$ and $\tau = 0$; the smectic-A filaments, which are characterized as plane curves, are on a plane perpendicular to $\boldsymbol{E}$) into Eqs. (6) and (7), the shape equations (Eqs. (6) and (7)) are reduced to

$$k_c(\kappa^3 + 2\ddot{\kappa}) - (\lambda + \Pi - \Upsilon E^2)\kappa = 0. \tag{8}$$

Equation (8) is the shape equation for two dimensional smectic-A filaments under electric-field application. We define the filament's tangential unit vector to be $\mathbf{t}(s) = (\cos\phi(s), \sin\phi(s), 0)$. Inserting this into Eq. (8) we know that ϕ is determined by

$$\dot{\phi}^2 = \frac{\lambda + \Pi - \Upsilon E^2}{k_c} + \frac{f}{k_c}\cos(\phi - \phi_0), \tag{9}$$

where f is a positive constant, ϕ_0 is an integration constant, and $\dot{\phi} = \mathrm{d}\phi/\mathrm{d}s$ [5]. Figure 3 shows the solution to Eq.(9) which is a periodic function. The parameter used in the calculation were: ρ_o=2.0 μm, ρ_i=0.4 μm, γ=10 pN/μm, E=0.14 V/μm, $\Delta\varepsilon$=9, k_{11}=10 pN, g_0=16$\times$10^{-6} pJ/μm^3, f=0.4 pN, and the values of γ, k_{11}, g_0 and f are reasonable in a smectic-A



$E = 0.14\ \mathrm{V}/\mu\mathrm{m}$

FIGURE 3 A solution to the shape equation for a two-dimensional filament under electric-field application. The bar indicates 40 μm.

phase in an isotropic phase [1,3,5-7]. This solution is in qualitative agreement with the experimental result shown in Fig. 2(b). We calculated the solutions to Eq. (9) as a function of electric field, and found that the experimental observation shown in Figs. 2(b)-2(d) (the amplitude of periodic smectic-A filaments is decreased and the periodicity along the growth direction is increased with increasing electric field) can be qualitatively reproduced.

CONCLUSIONS

We have studied the effect of electric-field application on the pattern formation of smectic-A filaments in an isotropic phase. When the LC samples are subjected to electric field of 0.14 V/μm, the three dimensional pattern of smectic-A filaments is drastically changed to the two dimensional pattern of smectic-A filaments, which is formed on a plane perpendicular to electric field. The electric field-induced two-dimensional pattern of smectic-A filaments is a periodically buckled structure, and the amplitude of the periodically buckled filaments is decreased and their periodicity is increased with increasing electric field. It is found that the periodically buckled filaments are equilibrium shapes. We have analyzed these experimental findings in terms of analytical approach to the variation problem for equilibrium shapes of smectic-A filaments in an isotropic phase. We derive the shape equations by a variation of the total energy consisting of F_V, F_A, F_C and F_D. We have shown that the experimental findings can successfully be explained; a periodically buckled filament is a solution to the shape equations and the electric-field dependence of the amplitude and the periodicity of the buckled filaments are well reproduced.

Acknowledgement

We would like to thank financial support by the Seki Memorial Foundation for the Promotion of Science and Technology and by the Sasakawa Scientific Research Grant from The Japan Science Society.

References

[1] H. Naito, M. Okuda and Z.C. Ou-Yang, *Phys. Rev. E* **55**, 1655 (1997); W. E and P. Palffy-Muhoray, *J. Nonlinear Sci.* **9**, 417 (1999).
[2] A. Adamczyk, *Mol. Cryst. Liq. Cryst.* **170**, 53 (1989); A. Adamczyk, *Mol. Cryst. Liq. Cryst.* **261**, 271 (1995).
[3] H. Naito, M. Okuda and Z. C. Ou-Yang, *Phys. Rev. Lett.* **70**, 2912 (1993).
[4] H. Naito, M. Todorokihara, Y. Hirose and Z. C. Ou-Yang, *J. Phys. Soc. JPN.* **67**, 713 (1998); M. Todorokihara, H. Naito and Z. C. Ou-Yang, *Mol. Cryst. Liq. Cryst.* **328**, 549 (1999).
[5] H. Zhou and Z. C. Ou-Yang, *Phys. Rev. E* **58**, 5909 (1998).
[6] R. Pratibha and N. V. Madhusdana, *J. Phys. II France* **2**, 383 (1992).
[7] S. L. Arora, P. Palffy-Muhoray and R. A. Vora, *Liq. Cryst.* **5**, 133 (1989); P. Palffy-Muhoray, March 1989 meeting of the APS, St. Louis, MO, *APS Bulletin* **34**, 495 (1989); *Science News* March 25, 1989; P. Palffy-Muhoray *et al.*, *J. Phys. II France* **2**, 383 (1992).