Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

On Discrete Soliton and Soliton Lattice at SmC_{α}*-SmC Transition Driven by an Electric Field

Yuki Sakai [a], Masashi Torikai [a] & Mamoru Yamashita [a]
[a] Department of Physics Engineering, Mie University, Kurimamachiya, Tsu, Japan

Version of record first published: 22 Feb 2010

Mol. Cryst. Liq. Cryst., Vol. 516: pp. 81–90, 2010

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400903400688

On Discrete Soliton and Soliton Lattice at SmC$^*_\alpha$-SmC Transition Driven by an Electric Field

YUKI SAKAI, MASASHI TORIKAI,
AND MAMORU YAMASHITA

Department of Physics Engineering, Mie University, Kurimamachiya,
Tsu, Japan

Unwinding process of Smectic C^*_α phase (SmC^*_α) to smectic C phase (SmC) in an electric field looks similar to a transition from chiral smectic C phase (SmC^*) to SmC which is interpreted as a soliton condensation. As a pitch of the helical structure is quite short in the former, a discrete description is required and the soliton accompanying with should be a discrete type, while in the latter, the soliton is a kink of sine-Gordon equation. Under the condition of constant tilt at SmC^*_α, it has been elucidated that for the helical pitch larger than four-layer the transition is second order and a wave number versus field relation makes a devil's staircase. Here, a free energy curve for the wave number is proved to be non-differentiable at any rational wave number, monotonous and convex, corresponding to the devil's staircase structure of the wave number. This non-analytic property contrasts with the case of continuous description in SmC^*, where the free energy curve is analytic. In the framework of the present model, a change of apparent optical axis and switching current is calculated, which are compared with experimental results reported so far.

Keywords Devil's staircase; discrete soliton; non-differentiability of free energy; $SmC_\alpha{}^*$-SmC transition; switching current

1. Introduction

In some antiferroelectric smectic materials, various intermediate phases appear between smectic A phase (abbreviated SmA) and antiferroelectric phase, assigned as SmC$^*_A$. Chiral smectic C_α phase (SmC$^*_\alpha$) is one of such intermediate phases appearing just below SmA. This phase is ferroelectric and has a helical structure like chiral smectic C phase (SmC*) [1–3]. In both phases, the helical structures are unwound due to a coupling of polarization to an electric field applied in a direction perpendicular to the helical axis, and phase transitions to smectic C phase (SmC) occur [3–7].

The authors would like to thank Professor Atsuo Fukuda for his invaluable discussions. One of the authors (M. Y.) also thanks Professor Kazuo Sasaki for notifying him of Ref [16].
Address correspondence to Mamoru Yamashita, Department of Physics Engineering, Mie University, Kurimamachiya 1577, Tsu 514-8507, Japan. E-mail: mayam@phen.mie-u.ac.jp

In SmC*, a helical pitch is very large and the phase change is described in a continuum expression for a free energy, and the transition between SmC* and SmC is interpreted as a soliton condensation of sine-Gordon equation [8,9]. This transition is a typical example of a transition of nucleation type in contrast with an instability type of phase transition [10]. An excitation energy of soliton vanishes at a critical field, and just below the critical field, many solitons may be excited. However, in case of a repulsive interaction between solitons, those are located with equal separation having finite density, achieving a periodic structure called soliton lattice. On the other hand in SmC^*_α, a pitch of the helical structure at vanishing field is very small [11,12], and accordingly a discrete description is required. Then, the soliton should be replaced by one of discrete type, and it is interesting how properties of the phase transition to SmC and of the soliton change [13,14].

In the previous report, it is elucidated that branching of a response to the electric field occurs at three-layer structure [14]. For a pitch larger than four layers, the wave number versus field relation makes a devil's staircase structure [14–16] and the wave number vanishes continuously at the critical field, showing that the interaction between solitons is repulsive. On the other hand for the pitch smaller than three, the phase changes to bi-layer structure, which eventually transforms to the uniform SmC continuously. The branching occurring at three-layer structure is interpreted to correspond to an experimental evidence on birefringence change in the electric field in MHPOCBC reported recently by Shtykov *et al.* [17]. The theoretical results above-mentioned are derived under a condition of constant tilt angle. The model we have used was already studied in detail by Yokoi *et al.* [16], where comprehensive phase diagram together with soliton property are reported. Here, we report some facts derived newly for the response of SmC^*_α to the electric field in the framework of the constant tilt.

In the next section, formalism is introduced and discrete soliton solution is obtained numerically, from which an excitation energy curve of soliton for the electric field is derived. Following to this, it is proved that a wave number dependence of free energy density is non-differentiable at any rational wave number, corresponding to the devil's structure of the wave number versus electric field profile. In section 4, an apparent optical axis and switching current is calculated at the present model and compared with experimental results reported so far. Finally, brief summary is given together with an appendix where SmC*-SmC transition is reviewed.

2. Discrete Formslidm for SmC^*_α and Discrete Soliton

Under the condition of constant tilt angle, θ_0, a free energy for SmC^*_α is given in a discretized form by [14,15,18]

$$F = -\sum_i [K\theta_0^2 \cos(\varphi_{i+1} - \varphi_i - \delta) + E\theta_0 \cos\varphi_i], \tag{1}$$

in which φ_i denotes an azimuthal angle of direction of an electric polarization at i-th layer, to which the tilt c-director makes a right angle, δ a wave number at vanishing field in a scale unit of layer thickness d, K an elastic constant and an electric field E is applied in x-direction. This type of free energy is a direct generalization of the one for SmC*, given by Eq. (A1) in appendix [18], and already used at a study of helicity unwinding at antiferroelectric smectic C phase [19]. Minimum condition of F leads to the following equation,

$$\sin(\varphi_{i+1} - \varphi_i - \delta) - \sin(\varphi_i - \varphi_{i-1} - \delta) - e\sin\varphi_i = 0, \tag{2}$$

where $e = E/K\theta_0$. Equation (2) is analyzed numerically.

First, we obtain a soliton solution to Eq. (2). In the comprehensive study by Yokoi *et al.* [16], soliton properties are discussed in detail, where the interaction between solitons is shown to be repulsive for the pitch larger than four layers. Here soliton solutin is obtained in a rather naïve method. Equation (2) is rewritten as a successive equation,

$$\varphi_{i+1} = \varphi_i + \delta + \sin^{-1}\{\sin(\varphi_i - \varphi_{i-1} - \delta) + e\sin\varphi_i\}. \tag{3}$$

From a symmetry property of the soliton at the centre, we take $\varphi_0 = \pi - \varphi_1$, where a trial value for φ_1 is chosen suitably. Then, we obtain φ_2, φ_3, $\varphi_4, \ldots$, successively. We adjust the value φ_1 so as φ_n converges to 2π for large n. In Figure 1, a profile of the soliton is shown, where $\delta/2\pi = 1/8$ and $e = 0.36725$. Excitation energy of one soliton, ΔF, is calculated simultaneously by the following equation,

$$\Delta F = \sum_i [-K\theta_0^2\{\cos(\varphi_{i+1} - \varphi_i - \delta) + e\cos\varphi_i\} - f_C], \tag{4}$$

where f_c is free energy per layer at SmC, $fc = -K\theta_0^2\ (\cos\delta + e)$. For various values of e, solitons are obtained, and values of the excitation energy are calculated. In Figure 2, field dependence of ΔF is shown for $\delta/2\pi = 1/8$. Needless to say, a field value at which ΔF vanishes coincides with the critical field, e_c, estimated from a periodic solution with quite large period [14]. For the field smaller than the critical field, ΔF is negative and many solitons may be excited, while because of repulsive interaction between solitons density of soliton remains finite, making the soliton lattice, *i.e.*, a periodic structure.

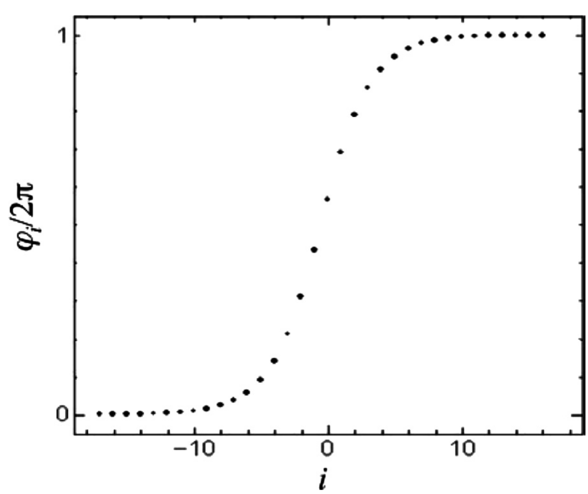


Figure 1. A profile of discrete soliton, where $\delta/2\pi = 1/8$.

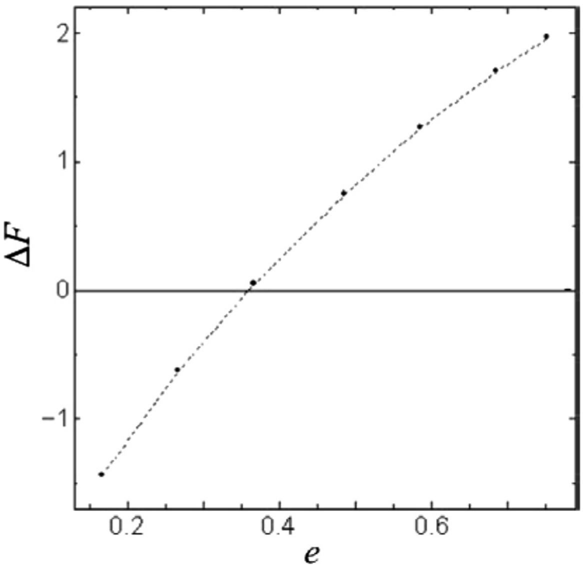


Figure 2. Excitation energy of discrete soliton, where $\delta/2\pi = 1/8$.

3. Non-Differentiability of Free Energy

Apart from the soliton solution, we have periodic solutions to Eq. (2), called soliton lattice. We consider the form, $\varphi_{i+p} = \varphi_i + 2\pi m$, ($i = 1, 2, \ldots, p$, m ($<p$) $= 1, 2, \ldots$) with period p in the scale d, and the wave number q given by $2\pi m/p$. It is noticed that $q/2\pi$ ($=m/p$) is nothing but soliton density [20]. In the previous study, q-e relation, $q(e)$, is shown to make a devil's staircase [14,16]. In case of continuous description in SmC*, free energy density is derived as an analytic function of the wave number given by Eq. (A1) using Eqs. (A2) and (A4), and corresponding to this, the wave number is a continuous function of the field as determined by Eqs. (A4) and (A5). Here, we show that the free energy as a function of the wave number is non-differentiable at any rational wave number.

A global profile of the free energy per layer, $f(q)$, is shown in Figure 3 at $\delta/2\pi = 1/4$ for various filed, where difference of $f(q)$ from f_c, $\Delta f = f(q) - f_c$, is depicted. Linear dependence near the origin is noticed [13,20]. At $e = e_c$, a tangent of Δf vanishes at the origin. The value of Δf comes from the interaction between solitons The relation of Δf to q in Figure 3 looks a smooth curve for each e-value. However, taken values of q are too rough to clarify the detail of $f(q)$. To see details of the profile, we choose wave numbers $q/2\pi$ in Farey series $n/(5n-1)$ and $n/(5n+1)$ with $n = 2, 3, 4, \ldots$, converging the $1/5$. The curve $f(q)$ is shown in Figure 4. The value of the field e is $e_c^{(-)}$ ($=1.044$) in (a), 1.130 in (b) and $e_c^{(+)}$ ($=1.215$) in (c), where the phase with $q/2\pi = 1/5$ is stable in an interval, $e_c^{(-)} < e < e_c^{(+)}$ [14]. Thus right derivative and left derivative are apparently different, and roughly speaking the difference between both derivatives is proportional to a width of stable region (in this case, $(e_c^{(+)} - e_c^{(-)})$). In Figure 4(a), the right derivative vanishes, showing a coexistence between phases of wave number $q/2\pi = 1/5$ and $n/(5n-1)$ with infinite n value. On the other hand in Figure 3(c), the coexistence between phases of wave number $1/5$ and $n/(5n+1)$ with infinite n value occurs. The f-q relation looks almost linear at both side of wave

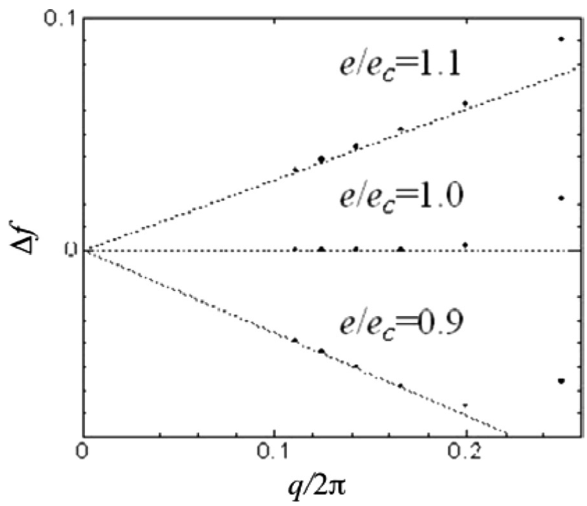


Figure 3. Wave number dependence of free energy per layer, Δf, where $\delta/2\pi = 1/4$.

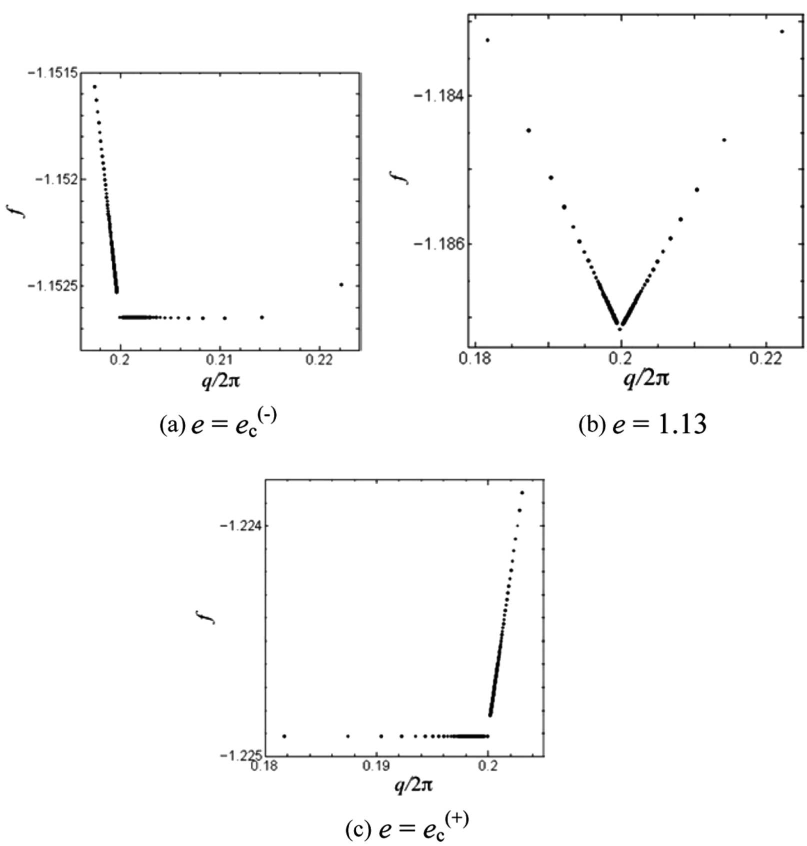


Figure 4. The $f(q)$ showing non-differentiability of Free energy at $q/2\pi = 1/5$.

number, $q/2\pi = 1/5$, because of shortness of the repulsive interaction between solitons as shown in Figure 3, where the linear dependence of $f(q)$ extends to about $q/2\pi = 1/5$ near the critical field. Because of the devil's staircase for $q(e)$, any state with rational q value is stable in a finite interval of e. Thus, we can show non-differentiability of $f(q)$ at any rational q value within a limit of numerical precision. Anyway, $f(q)$ is considered to be monotonous and convex, which correspond to the devil's staircase.

4. Optical Axis and Switching Current

Experimental observations of physical property of SmC^*_α have reported, not so much, so far, among which change of optical axis due to applied field [1] and switching current [2] are of interest. Here, we reproduce them in the framework of the present model.

In chiral ferroelectric smectics, director makes a right angle to the electric polarization. Assume an azimuthal angle of director $\boldsymbol{n}_i$ at the i-th layer, $\phi_i = \varphi_i + \pi/2$. Then, under the condition of constant tilt angle θ_0, $\boldsymbol{n}_i$ is given by ($\sin\theta_0 \cos(\varphi_i + \pi/2)$, $\sin\theta_0 \sin(\varphi_i + \pi/2)$, $\cos\theta_0$). By averaging $\boldsymbol{n}_i$ over one period, $<\boldsymbol{n}_i>$, we obtain

$$<\boldsymbol{n}_i>= (0, \sin\theta_0 < \cos\phi_i >, \cos\theta_0), \tag{5}$$

where $<\cos\varphi_i>$ is an average of $\cos\varphi_i$ over the period. Thus, an angle of the optical axis making θ from z-axis is given by

$$\tan\theta = \tan\theta_0 <\cos\phi_i>. \tag{6}$$

The tilt angle θ_0 in the present study is taken arbitrary, and here the average $<\cos\varphi_i>$ is chosen as a quantity to describe the change of the optical axis. In Figure 5, field dependence of $<\cos\varphi_i>$ $(=\Theta)$ is shown for $\delta/2\pi = 1/4$ (a) and 1/8 (b), in which wave numbers having relatively large stable regions are taken into account. Corresponding to the devil's staircase structure of $q(e)$, the change of

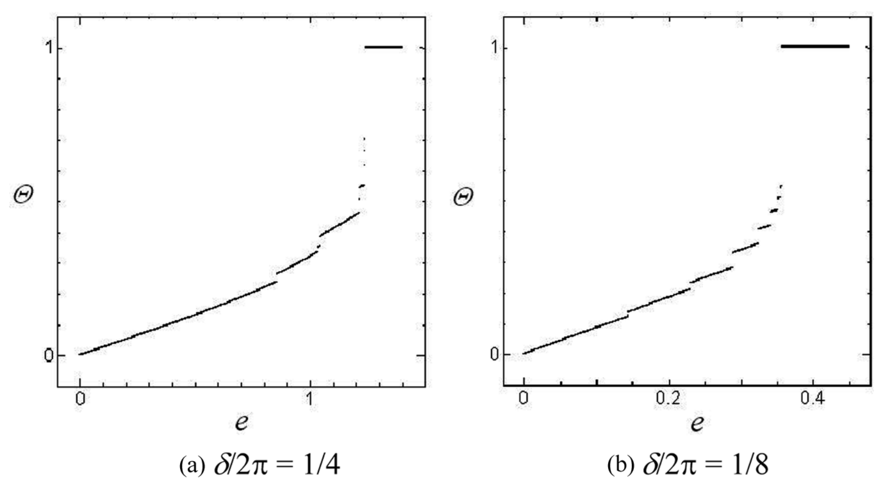
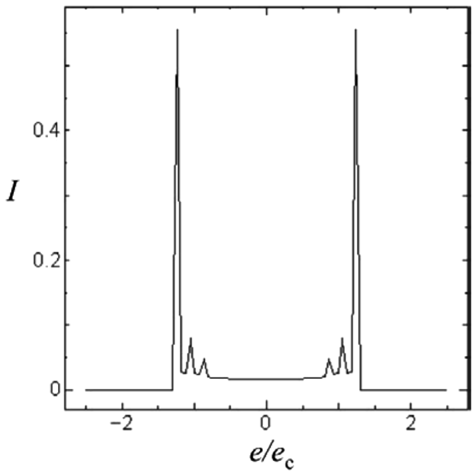
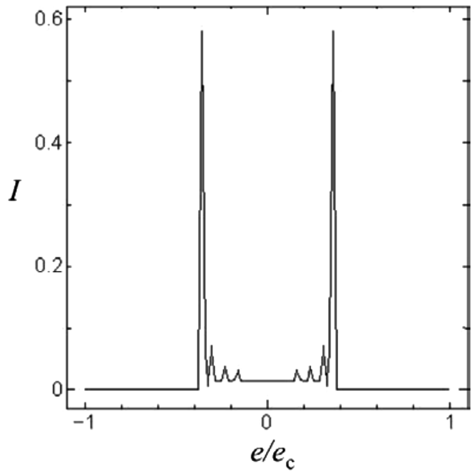


Figure 5. Electric field dependence of tilt angle.



(a) $\delta/2\pi = 1/4$



(b) $\delta/2\pi = 1/8$

Figure 6. Difference ratio I which is proportional to witching current at triangle wave.

Θ shows a staircase structure, which is compared with the experimental result in Ref. [1], where staircase behaviours are observed at SmC_{α}^{*}.

The electric polarization in x-axis is proportional to $\sin\theta_0 < \cos\varphi_i >$. The switching current comes from a change of the polarization. In an alternating electric field of triangle wave, a magnitude of the electric field is proportional to the time at an increasing field in one period. Accordingly, the switching current, i, is proportional to a derivative of Θ with respect to e, that is,

$$i \propto \frac{d\Theta}{de}. \tag{7}$$

The curves of Θ in Figure 5 are non-differentiable, and accordingly we replace the derivative in Eq. (7) by finite difference ratio. In Figure 6, I ($=\Delta\Theta/\Delta e$) is shown for $\delta/2\pi = 1/4$ and $1/8$, where we take $\Delta e = e_c/20$ and e-value is extended to negative by mirror imaging and I is scaled in an arbitrary unit. These curves are compared with the experimental results [2]. In the present stage, it is difficult to discuss qualitatively. However, profiles shown in Figures 5 and 6 are similar qualitatively to the experimental curves.

5. Summary

A discrete model as a generalization of sine-Gordon system describing unwinding process of chiral smectic C phase (SmC^{*}) is applied to chiral smectic C_{α} phase (SmC_{α}^{*}) in an electric field under a condition of constant tilt angle. Though the model has already been studied extensively [16], here we show a remarkable property of free energy as a function of wave number, that is, the free energy is non-differential at any rational wave number. This corresponds to a devil's staircase structure of the wave number versus field relation. In the framework of the present model with constant tilt angle, a change of optical axis for increasing field and switching current at applied electric field of triangular ware are calculated. These behaviours are in agreement qualitatively with the experimental results obtained so far [1,2].

In an antiferroelectric smectic materials, successive transition occurs [21]. To describe the successive phase transition, models with long range interactions have been introduced [22,23], while the present model is quite simple with only nearest layer interaction. Here, we are interested in an unwinding process of the helical structure not in the successive phase transition, and we consider the present model is suitable for the phenomena concerned.

In a continuum description for SmC*, transition to smectic C phase is continuous so long as a change of the tilt angle is neglected. On the other hand in the discretized model with interaction of cosine form, the first order transition is shown to occur for quite short period of helical structure shorter than four layers [16]. Anyway, the effect of the variation of the tilt angle is of interest, because the tilt angle is generally small in SmC_α^* and the variation of the tilt angle may not be neglected. This is an open problem in a future.

Appendix. Formalisms and Soliton Excitation in SmC*

Unwinding process of SmC* by the electric field is reviewed [8,9]. Under the condition of constant tilt angle $\theta(z) = \theta_0$, the Ginzburg-Landau free energy is given with the electric field $\boldsymbol{E}$ in x-axis by [18]

$$F = \int \left[\frac{1}{2} k\theta_0^2 \left(\frac{d\varphi}{dz} - q_0 \right)^2 - E\theta_0 \cos\varphi \right] dz, \tag{A1}$$

where φ $(=\varphi(z))$ denotes an azimuthal angle describing a direction of electric polarization at z, q_0 a wave number at vanishing field, k an elastic constant and a magnitude of the polarization is taken to be unity for simplicity. Euler-Lagrange equation for Eq. (A1) is given by

$$\frac{\mathrm{d}^2\varphi}{\mathrm{d}z^2} = \frac{E}{k\theta_0} \sin\varphi, \tag{A2}$$

which is nothing but the static sine-Gordon equation. A periodic solution to Eq. (A2) is obtained as

$$\varphi = 2\sin^{-1} \mathrm{sn}\left(\frac{1}{\kappa} \sqrt{\frac{E}{k\theta_0}} (z - z_0);\ \kappa \right) + \pi, \tag{A3}$$

in which $\mathrm{sn}(x,\ \kappa)$ denotes a Jacobi's sn-function with κ, a modulus. The state described by Eq. (A3) is called soliton lattice [8,9]. A wave number of the structure is derived as

$$q = \frac{\pi^2}{4} \frac{q_0}{K(\kappa)E(\kappa)}, \tag{A4}$$

where $K(\kappa)$ and $E(\kappa)$ are complete elliptic integrals of the first and second kinds, respectively, and the modulus κ is determined from the minimum condition of the free energy (A1) as a function of E by following relation,

$$\frac{\kappa}{E(\kappa)} = \frac{4}{\pi}\sqrt{\frac{E}{k\theta_0 q_0^2}}. \tag{A5}$$

Thus, q is given as a function of E by the use of Eq. (A5), showing a continuous curve. At a certain field strength, corresponding to the limit $\kappa = 1$, q vanishes in Eq. (A4), indicating a critical point, and from Eq. (A5) we obtain a critical field E_c as

$$E_c = \frac{\pi^2}{16} k\theta_0 q_0^2. \tag{A6}$$

At $\kappa = 1$, we have one-soliton solution to Eq. (A2), a kink, given by,

$$\varphi = 4\tan^{-1}\tanh\left\{\frac{1}{2}\sqrt{\frac{E}{k\theta_0}}(z - z_0)\right\} + \pi \tag{A7}$$

An excitation energy of the soliton is calculated as

$$\Delta F = 4\sqrt{\pi} k\theta_0^2 q_0 \left(\sqrt{\frac{E}{E_c}} - 1\right) \propto \frac{E - E_c}{E_c} \tag{A8}$$

The excitation energy vanishes at $E = E_c$, and For negative value at $E < E_c$, soliton lattice described by Eq. (A2) is achieved due to a repulsive interaction between solitons [8,9]. The relation $\Delta F(E)$ in Eq. (A8) corresponds to Figure 2 for discrete soliton. Thus, the transition between SmC* and SmC is interpreted as soliton condensation [8,9].

References

[1] Hiraoka, H., Takanishi, Y., Skarp, K., Takezoe, H., & Fukuda, A. (1991). *Jpn. J. Appl. Phys.*, *30*, L1819.

[2] Takanishi, Y., Takezoe, H., Fukuda, A., Isozaki, T., Suzuki, Y., & Kawamura, I. (1991). *Jpn. J. Appl. Phys.*, *30*, 2023.

[3] Hiraoka, H., Takanishi, Y., Takezoe, H., Fukuda, A., Isozaki, T., Suzuki, Y., & Kawamura, I. (1992). *Jpn. J. Appl. Phys.*, *31*, 3394.

[4] Bourny, V. & Orihara, H. (2001). *Phys. Rev.*, *E63*, 021703.

[5] Orihara, H., Naruse, Y., Yagyu, M., Fajar, A., & Uto, S. (2005). *Phys. Rev.*, *E72*, 040701.

[6] Meyer, R. B., Liebert, L., Strzelecki, L., & Keller, P. (1975). *J. Phys. Lett. France*, *36*, L-69.

[7] Meyer, R. B. (1977). *Mol. Cryst. Liq. Cryst.*, *40*, 33.

[8] Yamashita, M. (1985). *Prog. Theor. Phys.*, *74*, 622.

[9] Yamashita, M. (1991). In: *Solitons in Liquid Crystals*, Lam, L. & Prost, J. (Eds.), Springer-Verlag: Berlin, Chap. 10.

[10] de Gennes, P. G. (1975). In: *Fluctuations, Instability and Phase Transition*, Riste, T. (Ed.), Plenum Press: New York & London, 1.

[11] Mach, P., Pindak, R., Levelut, A.-M., Barois, P., Nguyen, H. T., Huang, C. C., & Furenlid, L. (1998). *Phys. Rev. Lett.*, *81*, 1015.

[12] Mach, P., Pindak, R., Levelut, A.-M., Barois, P., Nguyen, H. T., Baltes, H., Hird, M., Toyne, K., Seed, A., Goodby, J. W., Huang, C. C., & Furenlid, L. (1999). *Phys. Rev. E*, *60*, 6793.
[13] Torikai, M. & Yamashita, M. (2007). *Mol. Cryst. Liq. Cryst.*, *465*, 239.
[14] Sugimoto, Y., Torikai, M., & Yamashita, M. (2009). *Mol. Cryst. Liq. Cryst.*, *511*, 348.
[15] Bak, P. & Bruinsma, R. (1982). *Phys. Rev. Lett.*, *49*, 249.
[16] Yokoi, C. S. O., Tang, L.-H., & Ghou, W. (1988). *Phys. Rev.*, *B37*, 2173.
[17] Shtykov, N. M., Chandani, A. D. L., Emelyanenko, A. V., Fukuda, A., & Vij, J. K. (2005). *Phys. Rev. E*, *71*, 021711.
[18] Pikin, S. A. & Indenbom, V. L. (1978). *Sov. Phys. –Usp.*, *21*, 487.
[19] Parry-Jones, L. A. & Elston, S. J. (2001). *Phys. Rev.*, *E63*, 050701(R).
[20] Yamashita, M., Kimura, H., & Nakano, H. (1981). *Mol. Cryst. Liq. Cryst.*, *68*, 79.
[21] Matsumoto, T., Fukuda, A., Johno, M., Motoyama, Y., Yui, T., Seomun, S. S., & Yamashita, M. (1999). *J. Mater. Chem.*, *9*, 2051.
[22] Cepic, M. & Zeks, B. (2007). *Mol. Cryst. Liq. Cryst.*, *236*, 61.
[23] Emelyanenko, A. V., Fukuda, A., & Vij, J. K. (2006). *Phys. Rev. E*, *74*, 011705.