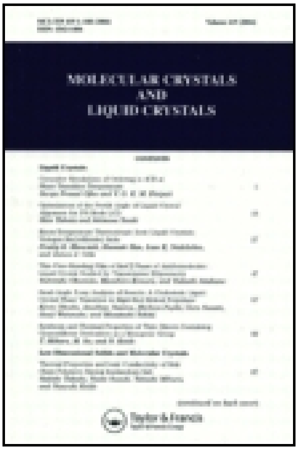

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Observation of One-Dimensional Spinodal Decomposition in a Nematic Liquid Crystal

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OBSERVATION OF ONE-DIMENSIONAL SPINODAL DECOMPOSITION IN A NEMATIC LIQUID CRYSTAL

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The coarsening dynamics of kinks in a one-dimensional conserved order parameter system is investigated experimentally in a nematic liquid crystal of negative dielectric anisotropy under a magnetic field. Without an external electric field, a straight splay-bend wall is formed in the cell perpendicular to the magnetic field. When an electric voltage, higher than a critical value, is applied to the liquid crystal cell, the straight wall spontaneously tilts locally toward the magnetic field direction and forms a zigzag line. Then the zigzag pattern starts to coarse. Considering the vertexes of the zigzag as kinks in a one-dimensional conserved Ising system, the coarsening dynamics is discussed. It is found that, as expected by the theories for one-dimensional kink dynamics, the characteristic length increases logarithmically in time and the dynamical scaling law holds in the coarsening process.

Keywords: kink dynamics; pattern formation; spinodal decomposition

INTRODUCTION

Phase ordering dynamics has been investigated intensively in the last two decades [1,2]. In the experimental field, Orihara and Ishibashi [3] were the first to use a nematic liquid crystal to investigate the phase ordering

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dynamics in the two-dimensional non-conservative Ising system. Following their work, a number of experimental studies [4,5] that utilize the properties of liquid crystals have been undertaken for various kinds of non-conserved order parameter systems and a lot of knowledge has been accumulated. However, probably due to experimental difficulties, there have not been any detailed experimental studies for the one-dimensional situation.

Recently, two similar experimental systems for studying an Ising conserved order parameter system in one-dimensional space have been proposed [6,7]. In the first case, 4-cyano-4′-pentyl biphenyl (5CB), whose dielectric anisotropy is positive $\Delta\varepsilon > 0$, was used [6]. In the second one, p-methoxybenzilidene-p-n-butylaniiliinee (MBBA), $\Delta\varepsilon < 0$, was used [7]. In both systems, a straight splay-bend wall, or so-called “Ising wall”, formed under a magnetic field is spontaneously transformed into a zigzag wall by application of a suitable electric voltage. Though the zigzag deformation increases the length of the wall and consequently tends to increase free energy, the twist deformation besides the wall, which was not present before the zigzag transition and replaces the initial slay-bend deformation, compensates for the increase of wall energy and reduces the total deformation energy of the system. This occurs as the twist deformation is more favorable than the other elastic deformation [8]. The neighboring zigzag vertexes attract each other and disappear by coalescence while the zigzag angle remains constant. As a result, the number of zigzags decreases and the average width of the zigzag increases with time. In the case of MBBA, due to the electrohydrodynamic instability, a pair of convective rolls is induced locally along the wall. Since the speed of coarsening in MBBA is much faster than that in 5CB, the hydrodynamics can probably be considered as a simple amplifier of the elastic anisotropy effect. Though there is no electrohydrodynamic convection in the case of 5CB, the explanation for the presence of a conserved quantity in the coarsening process is the same in both the experiments. It is derived from the geometrical constraints of the problem: the zigzag process is not associated with a global translation perpendicular to the initial straight wall, and it comes from the trivial conservation of the zig and zag projection length, along the initial straight wall. It is consequently very clear from the continuity of the wall and the conservation of zigzag angle, that the time evolution of the zigzag wall can be considered as a one-dimensional analog of spinodal decomposition. This consideration can be applicable for a zigzag instability in electrohydrodynamic convection (EHC) [8,9], which extends over a whole cell of nematic liquid crystal with negative dielectric anisotropy. Indeed, Sasa pointed that if the zigzag angle is uniform along a preferred direction of molecules, which is defined by a direction of external magnetic field or rubbing treatment, a coarsening of zigzag pattern perpendicular to the preferred direction can be considered as the spinodal decomposition in a one-dimensional conserved order parameter system [10]. In that case, the one dimensional Cahn-Hillard equation can be deduced from a model equation of EHC [10]. In the extended version of EHC, however, the uniformity of the zigzag angle along the preferred direction is not sufficient for the experimental study of the coarsening dynamics in one-dimensional conserved order parameter system.

The aim of this paper is to investigate experimentally the spinodal decomposition in the one-dimensional conserved order parameter system. For the sake of experimental convenience, we have considered the zigzag wall in MBBA [7]. In this paper, we discuss the features of coarsening in terms of a characteristic length, a spatial autocorrelation function of the Ising domains and a size distribution function of the domains. Moreover, the basic interaction between the kinks is estimated by analyzing the annihilation process of two neighboring kinks.

EXPERIMENT

The liquid crystal used is p-methoxybenzilidene-p'-n-butylaniline which is sandwiched between two glass plates coated with a homeotropic anchoring agent (Nissan Chemical Industries, SUNEVER SE-1211). The thickness and the size of the cell are $d = 50\,\mu m$ and $2.0\,cm \times 1.5\,cm$, respectively. As illustrated in Figure 1, the sandwiched cell is placed above two permanent Nd-Fe-B magnets that produced a slightly inhomogeneous magnetic field inside the cell. Below the magnets stage a polarizer is placed. In this experiment, the distance between the cell and the magnets is fixed. Though the inhomogeneous magnetic field exerts a restoring force on the Ising wall if the wall is moved from its equilibrium position, the strength of the restoring force is considered as negligible, the magnetic field being almost uniform and fixed 315 mT along the x-axis in the vicinity of the wall. The straight wall in the absence of an electric field is parallel to the y-axis.

The ac electric field is applied by a synthesizer (Hewlett Packard HP 3145A). The voltage and frequency are fixed as 8.0 Vr.m.s. and 20 Hz, respectively. The threshold voltage for zigzag instability is 6.9 Vr.m.s. A CCD camera (SONY XC-55CE) of CCIR standard mounted on a microscope (Olympus, BH-2) sends the image signal to a video capture board (Scion Corporation, LG-3). A sequence of images consisting of 12 snapshots with exponential time step based on 2s is stored in a personal computer (Apple, PowerMacintosh G4), where the images are digitized into 256 gray levels and 760×512 pixels. The images are analyzed by using NIH-Image (National Institute of Health in U.S.A. NIH-Image) with originally developed subprograms. The statistical quantities discussed in the present paper are evaluated from 10 runs of the same experiment.

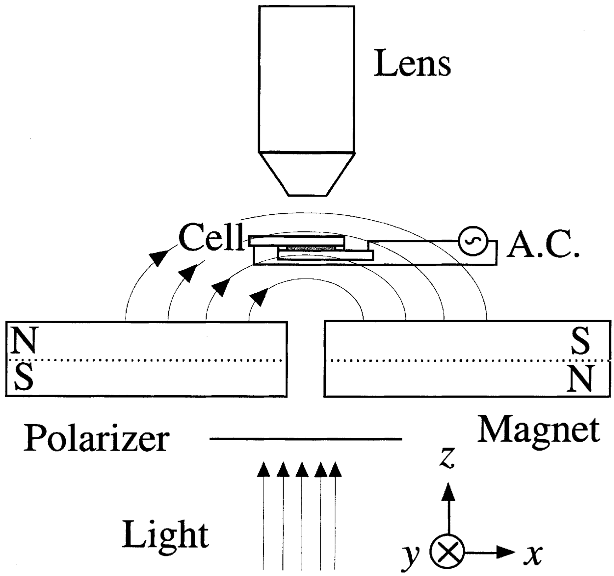


FIGURE 1 Schematic illustration of experimental system.

RESULTS AND DISCUSSIONS

The typical coarsening process of zigzag wall after the application of electric field is shown in Figure 2. The positions of vertexes can be estimated by applying the following image processing techniques. Firstly, a smoothing

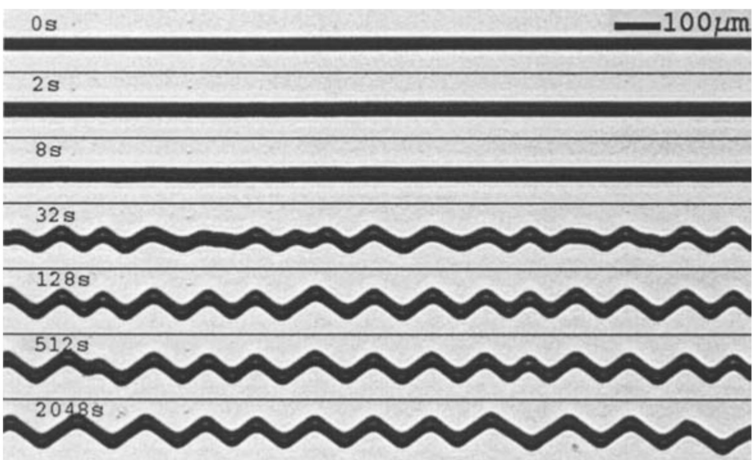


FIGURE 2 Coarsening process of zigzag pattern.

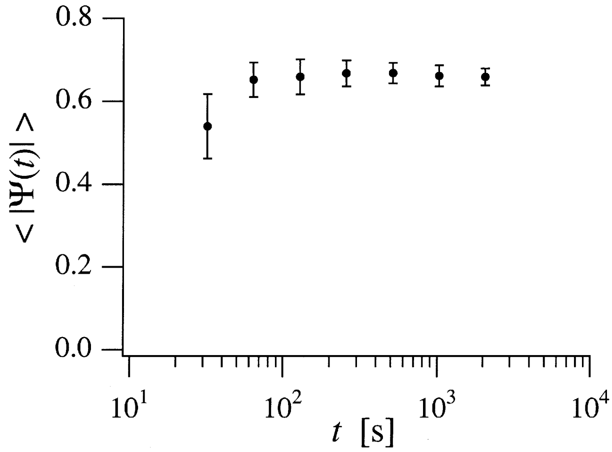


FIGURE 3 Time evolution of the average zigzag angle.

operation is undertaken to remove any short wavelength noise from the image and a binary image i.e. a black and white image, is created by setting a suitable intensity threshold. Next, the center of zigzag line is estimated. A least square fitting for each straight part of the zigzag line is then applied and a position and a local slope of zigzag are estimated.

The time evolution of the mean absolute values of the slope, $\langle|\Psi(y,t)|\rangle$, and the width, $W(t)$, are shown in Figure 3 and Figure 4, respectively. After $t \sim 64$s the average of zigzag angle becomes an equilibrium value and does not change at subsequent time. It is remarkable that $W(t)$, i.e. the

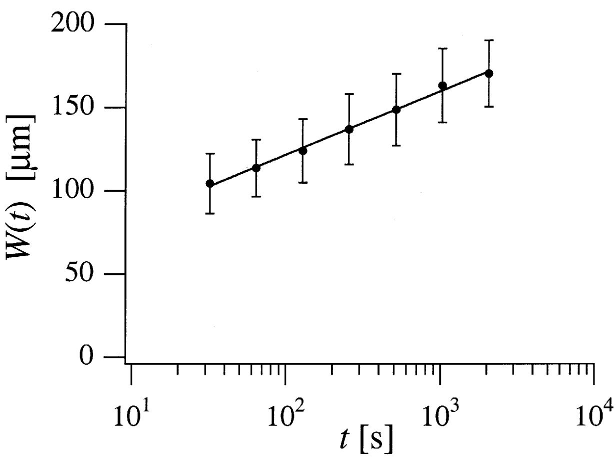


FIGURE 4 Logarithmic time evolution of the average width of zigzag.

characteristic length of this system, increases logarithmically in time, which agrees with the theories for the kink dynamics in the conserved order parameter system [11–14]. It should be recalled that in such theories [11–14] and numerical calculations [12], there is an attractive exponential interaction between the kinks.

In this coarsening process, two adjacent vertexes attract each other and the mutual distance between them decreases with time, demonstrating therefore, the presence of an attractive interaction between them. To estimate the interaction between the vertexes, the mutual distance between the annihilating vertexes is measured as a function of time. Three sets of time dependence of mutual distances are shown in Figure 5. Unfortunately, in contrast to the case of non-conserved order parameter system, it is impossible to construct a simple equation of motion for the mutual distance between two annihilating kinks in the present system, even if the interaction is known since their motion also depend on the position of other kinks [14]. It is probably the same reason which leads to the scattering of the data in Figure 5 especially in the early time regime. In all data Figure 5, the annihilation speed is accelerated drastically when their distance becomes around 30 μm which is a distance comparable with the width of the wall. This feature strongly suggests the existence of exponential attractive

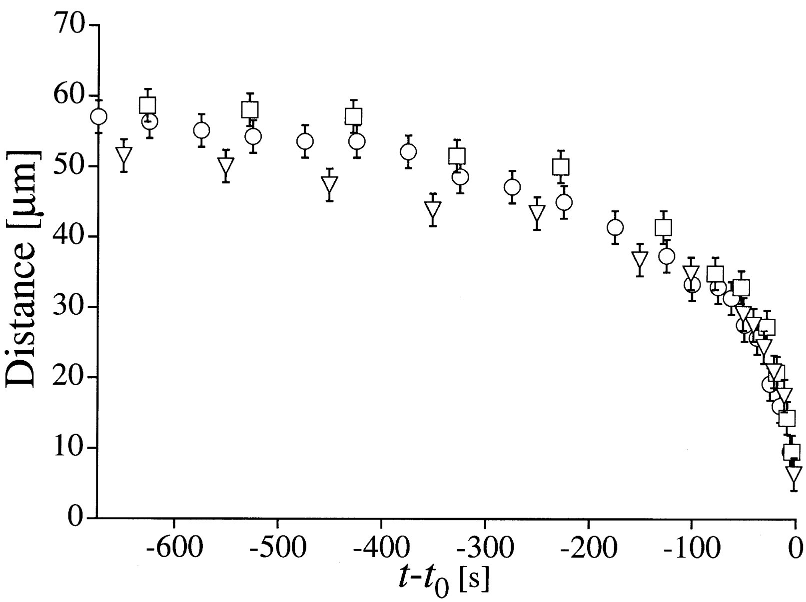


FIGURE 5 Mutual distance between two annihilating vertexes. The vertexes coalesce and disappear at $t = t_0$.

interaction between the vertexes as expected in the theories of the kink dynamics in one-dimensional system [11–14]. This estimation agrees with the logarithmic growth of the characteristic length.

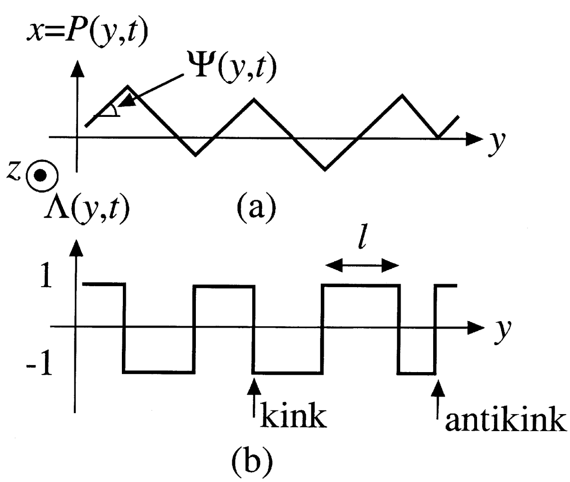
In order to discuss the statistical feature of spinodal decomposition, an Ising order parameter $\Lambda(y,t)$ defined as

$$\Lambda(y,t) = \begin{cases} 1 & \text{if } \Psi(y,t) < 0, \\ -1 & \text{if } \Psi(y,t) > 0, \end{cases} \tag{1}$$

is introduced. The conservation law is expressed as

$$M = \int \Lambda(y,t)dy, \tag{2}$$

where—is a time independent constant close to zero in the present case. Chevallard *et al.* [6] have derived this conservation law and the Cahn-Hillard type equation of motion for the zigzag wall in a nematics from the equation of director based of Frank's elastic free energy. Usually, the conservation law is associated to the conservation of irreducible physical quantities like a mass of component for the case of phase separation in binary alloy. But in the present case, it is caused by the continuity of the walls and the conservation of zigzag angle. Strictly speaking, the conservation law in the present system does not hold for the long time limit where there are only a few zigzags and the ends of the zigzag at the boundary of cell can move if the magnetic field is homogenous. Actually, however, the present system can be adequately regarded as a conserved order parameter system



Q1 **FIGURE 6** Schematic illustration of the relation between the zigzag and the kinks.

during the observation as long as the width of zigzag at $t = 2048$s remains less than one hundredth of the cell size.

The statistical features of the coarsening are analyzed by measuring the spatial autocorrelation function, $C(y,t)$, and the distribution function, $g(l,t)$, for domains separated by kinks. The phase ordering dynamics, $C(y,t)$ and $g(l,t)$ are defined

$$C(y,t) = \langle \Lambda(y,t)\Lambda(0,t) \rangle \tag{3}$$

and

$$g(l,t) = \frac{1}{N(t)} \sum_{i=1}^{N(t)} \delta(y_{i+1}(t) - y_i(t) - 1), \tag{4}$$

respectively, where $N(t)$ and y_i(t) denote the total number of kinks in the observed area and the position i-th kink at time t, respectively [12]. The probability that a domain is of a size in the interval $(l, l + dl)$ becomes $g(l,t)dl$. As shown in Figure 7, the correlation function shows an oscillation which is characteristic for conserved order parameter system. The location of first peaks at different times correspond to $W(t)$. As shown in Figure 8, $g(l,t)$ has a peak at a length $l \sim W$(t)/2. The cut-off length $l_c(t)$, which exists in the non-conserved order parameter system [12], also exists in the present system, below which $g(l,t)$ is very small. This means that domains with sizes smaller than $l_c(t)$ have been annihilated while sizes lager than $l_c(t)$ still survive. This result also supports the existence of strong short-range interaction like an exponential attractive force mentioned above. In addition, it should be noted that the shapes of $g(l,t)$ obtained

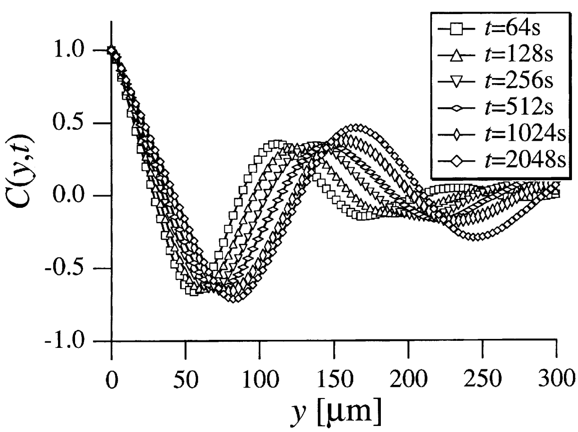


FIGURE 7 Autocorrelation function.

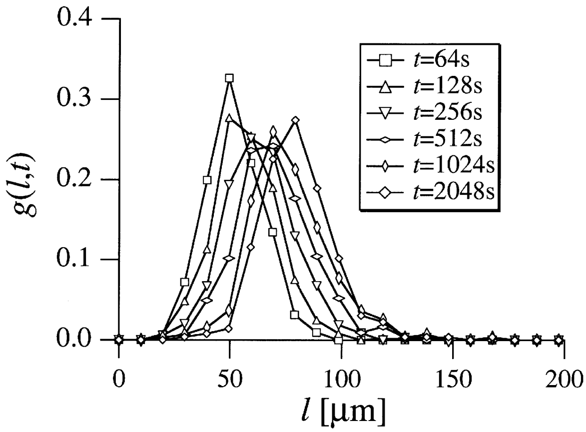


FIGURE 8 Size distribution function.

in the present experiment are not similar to that in the numerical simulation and the theoretical prediction for the non-conserved order parameter system by Kawasaki *et al.* [12] where the distribution of domain size has a very steep peak just above the cut-off length and very long tail over the peak. In the present system, as shown in Figure 2, the zigzag is relatively regular at late times, namely the domains are distributed narrowly, which seems to be a characteristic feature of conserved order parameter system.

Since the characteristic length shows logarithmic growth and the shapes of $C(y,t)$ and $g(l,t)$ at different times are very similar, it is easily expected that a dynamical scaling law holds in the coarsening process. To confirm

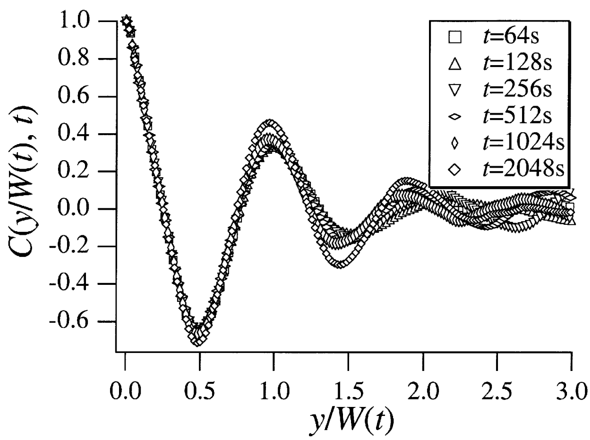


FIGURE 9 Scaled autocorrelation function.

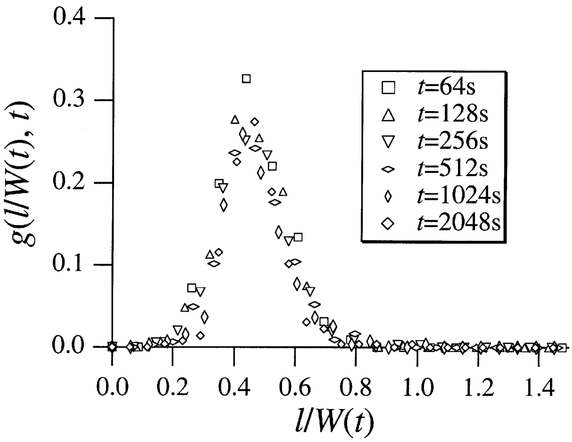


FIGURE 10 Scaled size distribution function.

this, spatial axes in $C(y,t)$ and $g(l,t)$ are scaled by $W(t)$ and re-plotted in Figure 9 and Figure 10, respectively. In each figure, the curves at different times fall on a universal curve, which shows clearly the existence of the dynamical scaling law.

SUMMARY

The coarsening of zigzag wall is investigated experimentally from the viewpoint of phase ordering dynamics. The characteristic length scale increases logarithmically in time. The spatial autocorrelation function shows oscillatory decay that is characteristic for the conserved order parameter system. From the scaling analysis of the autocorrelation function and the size distribution function of domains, the existence of dynamical scaling law is demonstrated. These features completely agree with theories within references [11–14]. The distribution function of domain has a cut-off length, below which the probability density of domain is very small. Unlike the case for the non-conserved order parameter system, the distribution function of domains has a peak above the cut-off length and does not have the long tail in the large distance regime.

REFERENCES

[1] Gunton, J. D., Miguel, M. S., & Sahni P. S. (1983). *Phase Transitions & Critical Phenomena* Domb C. & Lebowitz, J. L., (Eds.), Academic, New york: Vol. 8, 267.
[2] Bray, A. J. (1994). *Adv. Phys.*, *43*, 357.

[3] Orihara, H. & Ishibashi, Y. (1986). *J. Phys. Soc. Jpn.*, *55*, 2151.
[4] Nagaya, T., Orihara, H., & Ishibashi, Y. (1991). *J. Phys. Soc. Jpn.*, *60*, 1572 *ibid*. *61*, 3511 (1992); *ibid*. *64*, 78(1995); Orihara, H., Nagaya, T. & Ishibash, Y., *Formation, Dynamics & Statistics of Patterns 2*, (Eds.) Kawasaki K., Suzuki M., World Scientific 1993.
[5] Pargellis, A. N., Turok N., & Yurke B., (1997). *Phys. Rev. Lett. 67*, 1570; Pargellis, A. N., Mendez, J., Srinivasarao, M., & Yurke, B. (1996). *Phys. Rev. E*, *53*, R25, Chuang, I., Durrer, R., Turok, N., & Yurke, B. (1991). *Science 251*, 1336. Chuang, I., Turok, N. & Yurke, B. (1991). *Phys. Rev. Lett.*, *66*, 2472. Yurke, B., Pargellis, A. N., Chung, I., & Turke, N. (1992). *Physica B*, *178*, 56. Chung, I., Yurke, B., & Pargellis, A. N. (1993). *Phy. Rev. E*, *47*, 3343.
[6] Chevallard, C., Clerc, M., Coullet, P., & Gilli, J. M. (2000). *Eur. Phys. J. E 1*, 179. Chevallard, C., Nobili, M., & Gilli, J. M. (2001). *Liq. Cryst.*, *28*, 179.
[7] Nagaya, T. & Gilli, J. M. (2002). *Phys. Rev. E*, *65*, 51708. Nagaya, T., Gilli, J. M., & Chevallard, C. Proceedings of the conference “4ème Rencontres du Non linéaire 2001” (Eds.) Pomeau, I. & Ribotta, R. (2002). Non Linéaire Publications, Orsay.
[8] de Gennes, P. G. & Prost, J. (1993). *The Physics of Liquid Crystals*, Second edition, Oxford Science Publications, New york.
[9] Kramer, L. (1996). *Pattern Formation in Liquid Crystals* (Eds) by Buka, A. & Kramer, L. (1996). Springer-Verlag, New York, 221.
[10] Sasa, S. (1990). *Prog. Theor. Phys.*, *84*, 1009.
[11] Rutenberg, A. D. & Bray, A. J. (1995). *Phys. Rev.*, *E 51*, 5499.
[12] Nagai, T. & Kawasaki, K. (1983). Physica 20A, 587.*ibid*. *134A*, 483 (1986); Kawasaki, K. & Nagai, T. Physica *121A*, 175 (1983); Kawasaki, K., Ogawa, A., & Nagai, T. Physica *B149*, 97 (1988); Kawasaki, K. & Ohta, T. Physica *116A*, 573 (1982).
[13] Kawakatsu, T. & Munakata, T. (1985). *Prog. Theoret. Phys.*, *74*, 11.
[14] Majumdar, S. N. & Huse, D. H. (1995). *Phys. Rev.*, E *52*, 270.