Phase Separation in the Wake of Moving Fronts: Experiments and Simulations

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Abstract

The formation of regular precipitate stripes in the wake of moving chemical reaction-diffusion fronts is investigated. Experiments on the $\text{NaOH} + \text{CuCl}_2$ reaction in PVA hydrogel yield stripes parallel or slightly oblique to the front that supplies the precursor of the precipitate. The pattern formation was modeled by phase separation described by the Cahn-Hilliard Equation. Computer simulations reproduced the parallel and the oblique striping as well. Stripes perpendicular to the front are unstable and cannot be observed, in complete agreement with the experiments.

Pattern formation in the wake of quenching fronts has also been investigated computationally, and compared to the previous results. It has been shown that below a certain front speed stripes perpendicular to the front will appear. Moreover, they will bend so that their growing end to be kept perpendicular even if the front changes its direction. This result can be important in designing several nanotechnological and lithographical processes.

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I. INTRODUCTION

Recently, there is growing interest in spontaneous pattern forming processes that can yield regular microscopic textures. A widely studied example is phase separation, shown by various chemical and physical processes [4, 5, 8, 15, 19, 20]. Most of the experiments are concerned with initially homogeneous systems, where pattern formation starts after a temperature quench shifts the system into an unstable state. However, a new stream of research is being defined by studies on those processes where phase separation takes place in the wake of moving fronts.

Two mechanisms are known to yield spinodal phase separation behind traveling fronts. In the first, the concentration of the phase-separating compound lies between the spinodal points, but the temperature only drops below the critical value, required for the instability to occur, behind a quenching front. Alternatively, when the concentration is initially in the stable regime, a source front shifting it in between the spinodal points can switch the system into the unstable, pattern-forming range. Some important aspects on these mechanisms are listed as follows.

Computational studies on phase separation under directional quenching are presented in [9] and [18]. The phase separation was studied in the framework of the Cahn-Hilliard equation. At high velocities of the cooling front, irregular morphology (IM) emerged. At decreasing front velocities, stripes parallel to the quenching front, termed lamellar morphology (LM), and stripes perpendicular to the front, termed columnar morphology (CM) were found. Although little attention has been paid to the systematic investigation of the textures when the stripes were oblique to the front, we will consider these kind of patterns, and refer to them as oblique morphology (OM).

The examination of phase separation in the wake of source fronts has been motivated by the desire to set up a minimal model of the Liesegang phenomenon, in which a series of precipitate stripes emerge in the wake of the diffusion front of a reagent, referred to as the “outer electrolyte”, that penetrates into a hydrogel containing an “inner electrolyte” [3, 7]. Modeling the formation of the one-dimensional Liesegang patterns has been achieved by assuming that the reaction of the electrolytes yields an intermediary compound first, that separates into high and low density regions according to the Cahn-Hilliard equation [1, 2].

Several experimental results have been reported on two-dimensional striped structures
formed in the wake of filiform source fronts. Besides the classical Liesegang experiments, a great variety of such patterns have been found in the NaOH + CuCl\textsubscript{2} reaction in polyvinyl-alcohol (PVA) hydrogel medium. Parallel and oblique morphologies have also been observed in this reaction. However, up to this time, no mathematical models have been elaborated for describing these formations.

In this paper we investigate the formation of the microscopic, striped (“secondary”) patterns emerging in the NaOH + CuCl\textsubscript{2} reaction running in PVA hydrogel sheets. The main characteristics of the parallel and oblique morphologies observed in the experiments have been reproduced by computer simulations based on phase separation described by the Cahn-Hilliard equation.

II. EXPERIMENT

The NaOH + CuCl\textsubscript{2} reaction has been studied in a “Liesegang-like” setup, with NaOH as the outer electrolyte, and CuCl\textsubscript{2} as the inner electrolyte homogenized in PVA hydrogel. Details of the sample preparation are described in [12, 14]. The NaOH penetrates into the gel by diffusion, and its reaction with the inner electrolyte leads to a great variety of precipitate structures. These have been classified into “primary” and “secondary” patterns. In the following, the formation of the secondary, striped microscopic patterns is examined.

First, the reaction-diffusion front of the NaOH sweeps through the gel, which is assumed to be the source of a compound that phase separates into high-density (CuO precipitate) and low-density (free of precipitate) stripes. The CuO precipitate stripes are mostly parallel to the reaction-diffusion front, and in some cases oblique morphology has also been found. Although the source front cannot be observed by the naked eye, the oblique morphology can be identified by an intrinsic characteristic: In the case of the lamellar morphology, the stripes appear one by one along the source front, that is, their growth does not proceed via the elongation of their end points. In contrast, in the case of the oblique morphology, the stripes have growing endpoints in the wake of the source front, and they form an angle with the envelope of their terminal points.

The character of the striped microscopic patterns is correlated with the speed of the source front. Although the source front is not visible, it is followed by a sharp precipitation
front, where a blue-green compound is formed that shows no structure when investigated with an optical microscope [12]. This is referred to as the 'active border' of the blue-green precipitate, it can easily be observed even by the naked eye, and is likely to follow the shape and the speed of the source front. If the front velocity is between 0.6 \(-\) 0.9 \(\mu/s\), uniformly distributed colloidal precipitate forms ahead of the active border, but behind the source front. When the velocity is even smaller, the precipitate starts to show a pattern: a regular structure of parallel stripes of colloidal precipitate appears. Smaller front velocities lead to longer stripe wavelengths [12]. Note that in contrast to the hypotheses presented in [12], it is not believed any more that the active border is the source of the stripes' precursor.

The first emerging stripes will be parallel to the active border. When the source front does not change its shape and orientation, the subsequent stripes will form parallel to the previous stripes and the front as well, giving rise to a lamellar morphology. According to video microscope observations, a possible scenario for the formation of the oblique stripes is the following: When the source front suddenly changes its shape or orientation, \(e.g.\) as a result of an influx of the outer electrolyte from a novel direction, the newly formed stripes cannot follow the front’s altered orientation, but form more or less parallel to the previous ones. The newly formed stripes elongate only up to the limit of the region already visited by the source front, with the envelope of their growing endpoints being parallel to the actual position of the front (fig. 1).

III. MODELING THE PATTERN FORMATION IN THE WAKE OF SOURCE FRONTS

The process of phase separation, occurring in the wake of the \(NaOH + CuCl_2\) reaction-diffusion front, has been modeled by the Cahn-Hilliard equation with a Ginzburg-Landau free energy [8]. Although one of the equilibrium densities of this free energy is negative, and therefore, in our case unphysical, the equation can easily be rescaled to a form where both equilibrium densities are positive. However, for the sake of simplicity and without offending the physical content, we have used the free energy with minima at \(-1\) and \(+1\).

In order to focus our attention to the pattern formation, the reaction-diffusion system that produces the phase separating chemical has not been included in our model. In order to describe the source front, a Gaussian-type source term \(S(x, t; v)\) has been added to the
Cahn-Hilliard Equation:

\[
\frac{\partial c(x, y, t)}{\partial t} = -\Delta [c(x, y, t) - c(x, y, t)^3 + \epsilon \Delta c(x, y, t)] + S(x, y, t; v)
\] (1)

where

\[
S(x, y, t; v) = A \cdot \exp \left[ -\alpha (ax + b - vt)^2 \right]; \quad (2)
\]

Initially, the concentration of the compound \(c\) is set to the stable magnitude \(c_0(x, y, 0) = -1 + \eta\) in the whole rectangular simulation area, where \(\eta\) is a random uniform deviate distributed between \(\pm 0.001\) [18]. This deviate has been added to the model in order to make it more realistic.

The value \(c_0\) is increased by the source, moving with constant speed \(v\), to the constant value \(c_f\) [10]. The speed \(v\) of the source, as well as the concentration \(c_f\) next to the source front, are considered as independent simulation parameters. Having the speed \(v\) fixed, the value of \(c_f\) is determined by the amplitude \(A\) and the width \(\alpha\) of the Gaussian source. If \(c_f\) lies in between the spinodal points, that is, \(-1/\sqrt{3} < c_f < 1/\sqrt{3}\), the system will be unstable against linear perturbations, and phase separation will take place in the wake of the front. As time goes on, the concentration profile \(c(x, y, t)\) tends to reach the equilibrium values, and a “ripening” of the regions with the stable concentrations will take place as well. However, the initial conditions, as well as the movement of the Gaussian-type source ”front” will strongly affect the emerging patterns. These features will be our primary concern.

The equation (1) was solved on a rectangular grid using the finite difference method [22]. Periodic boundary conditions have been used in both directions.

The time evolution of the system was computed by explicit simple time marching on a rectangular grid. The mesh size was \(\Delta x = \Delta y = 1\), while the time step was \(\Delta t = 0.01\). A negligible change in some selected simulation results was only observed when the mesh size was halved and the time step was diminished 10 times. It is also important to mention that the effect of the grid anisotropy on the simulation results was also of minor importance. This has been checked by comparing pattern formation in the wake of source fronts with different orientations.
IV. SIMULATION RESULTS: SOURCE FRONTS TRAVELING WITH CONSTANT SPEED

The speed of the source front, as well as the initial conditions, play a decisive role in determining the character of the pattern formation. In this section, our concern will be to investigate their influence.

Initially, the concentration was set to \( c_0(x, y, 0) = -1 + \eta \) in the simulation area. This was increased by the front to \( c_f \), a value being in between the spinodal points. In the following, the results for the case \( c_f = 0 \) will be presented, and major differences for \( c_f \neq 0 \) will be mentioned.

In our investigations, the source fronts were traveling with different constant speed values. Although in the experiments the front speed varies in time, this change is usually not significant for \( 5 \leq 10 \) stripe wavelength, and the front speed can be considered locally constant.

Different pre-patterns with \( c(x, y, 0) = 0 \) introduced in the \( x \in (5, 30) \) space units region of the simulation area highly affected the character of the patterns emerging even after the front sweeps through this region. Note that the source was started at \( x = 10 \) space units from the \( Y \)-axis of the simulation area.

In the following, patterning at three different initial conditions are presented. The effects of the front speeds are also discussed within these cases.

a.) In the simplest scenario, the front is started parallel to the \( Y \)-axis of the rectangular grid, and sweeps with constant speed and orientation toward the opposite edge. The concentration is \( c_0(x, y, 0) = -1 + \eta \) all over the simulation area, that is, no initial patterning is introduced in the system. Depending on the front speed, two characteristic morphologies were observed.

If the front speed is higher than \( v \approx 5 \), an irregular morphology builds up in the wake of the front. The explanation is straightforward: the relatively slow phase separation drops behind the rapidly progressing front. As a consequence, there will be a large unstable domain between the front and the region where phase separation occurs, and the scenario will essentially be the same as the phase separation in a field with a homogeneous concentration in the unstable regime.

At front speeds below \( v \approx 2 \), stripes parallel to the front are formed. This pattern is
referred to as a lamellar morphology. In the vicinity of the limit velocity, the stripes are staggered and coarse, but below \( v \approx 1 \), they become smooth and straight. Regular lamellar morphology appears at much lower front speeds as well, but the wavelength of the stripes is increased. This effect is reminescent of the results encountered in modeling the Liesegang phenomenon by the terms of a spinodal phase separation, namely the growing wavelength of the stripes in the wake of a source front with decreasing speed [2]. Note that the character of the patterns does not changed when \( c_f = \pm 0.12 \).

b.) In order to examine the stability of the lamellar morphology, the simulations have been performed such that randomly distributed spots with \( c(x, y, 0) = 0 \) were introduced in the \( x \in (5, 30) \) space units region of the simulation area. Despite the above random initial conditions, which disturb the first stripes that build up in the wake of the front, lamellar morphology, or oblique morphology, with a small angle, appears at \( v \in (0.5, 2) \). At \( v > 5 \) irregular morphology, and at \( v \approx 0.1 \) spotty irregular morphology appears in a 250x620 simulation area. The tendency to form the lamellar morphology slightly diminished when \( c_f = -0.12 \).

c.) In certain parameter regions, the form and orientation of the preceding stripes will strongly influence the location of the subsequent stripes. Since the growth of a stripe depletes its surrounding, the source front will recover in concentration necessary for the emergence of a new stripe only above a certain distance from the old one. In order to simulate this scenario, in the \( x \in (5, 30) \) space units region of the simulation area a regular structure of tilted stripes with \( c(x, y, 0) = 0 \) were introduced. The angle between the edge of the simulation area (the Y-axis) and the stripes was about 30 degrees, and the wavelength of the structure was about 8 space units. When the front sweeps through this “pre-patterned” region, its contribution will accumulate on the stripes with the unstable concentration \( c = 0 \), leading to a fast phase separation. In this way, a stable striped structure, oblique to the front, will develop. However, an oblique striped structure survives only around \( v \approx 1 \). At \( v \approx 2 \), slightly disturbed lamellar morphology formed. When \( v > 5 \) and \( v < 0.1 \), irregular morphology and spotty irregular morphology emerged in a 250x620 simulation area. No significant change was observed when \( c_f = \pm 0.12 \).
V. ROTATING SOURCE FRONTS

The oblique morphology in the NaOH + CuCl$_2$ reactions in PVA gel sheets usually appears when the traveling reaction-diffusion front changes its direction, while the newly forming stripes keep the orientation of previous stripes. This process was computationally modeled by a rotating source front segment in a simulation area of 1200x600 space units, having a length of 570 space units. The initial concentration in the whole simulation area was set to $c_0(x, y, 0) = -1 + \eta$. The source term added to the Cahn-Hilliard equation takes the form

$$S(x, y, t; \omega) = A \sqrt{(x-x_0)^2 + (y-y_0)^2} \cdot \exp \left\{ -\alpha \left[ (x-x_0)\sin(\omega t + \theta_0) + (y-y_0)\cos(\omega t + \theta_0) \right]^2 \right\}$$

The character of the pattern formation is a function of the front speed, that depends on the angular velocity, as well as the position along the radius. In our simulations, having a front length of 570 space units, striped patterns just behind the front appeared around the angular velocity interval $\omega \in (0.002 - 0.02)$. The dynamics of the pattern formation was as follows: The first stripe forms roughly along the initial position of the front. Although the orientation of the front is continuously altered, the newly formed stripes will “try” to form along the old ones, parallel with them. Since the front changes its orientation in the meanwhile, the above scenario will lead to an oblique morphology.

However, the simulations showed that the stripes cannot grow perpendicular to the front. Their elongation becomes unstable when the angle of the stripes formed to the front supplying the phase separating material reaches about 70 – 90 degrees. At this stage, in some domains just behind the front oblique stripes with a small angle appear. In some other domains irregular morphology appears. Later, the above scenario may repeat itself.

Note that in the vicinity of the outer endpoint of the rotating front, where the speed is relatively high, the source front may not immediately be followed by the phase separation. The outer core of the circular region will be patterned by a different mechanism, namely the striping initiated by the arc-like edge where the concentration changes from $c = 0$ to $c = -1$. This striping will start along the edge, and will spread inside the unstable region, until it meets the straight striping initiated by the front itself.

Finally we review the pattern formation at much higher and lower angular velocities.
When the angular velocity is higher than $\omega = 0.05$, the overwhelming majority of the phase separation takes place far behind the front. Two mechanisms play an important role in the pattern formation. As mentioned previously, a striping will be initiated by the arc-like edge, where the concentration changes from $c = 0$ to $c = -1$. However, in the inner regions, mostly irregular patterns will form. In the case of low angular velocities, when $\omega < 0.001$, spotty irregular morphology appears in the wake of the front with a length of 570 space units.

VI. QUENCHING FRONTS REVISITED

An alternative way to start the spinodal decomposition in the wake of a traveling front is to set the concentration of the phase separating compound $c$ in between the spinodal points, while the temperature is dropped below the critical value only behind the front. The quenching in our simulations has been realized by changing the sign of the second-order term in the Cahn-Hilliard equation.

Pattern formation in the wake of quenching fronts has been studied by computational analyzes similar to those performed in the case of source fronts. The main difference with respect to the pattern formation in the wake of the source fronts is the appearance of the columnar morphology at low front speeds, in agreement with Furukawa’s results \[9\]. With the concentration of the phase separating compound set to $c_0(x, y, 0) = 0$, the columnar morphology appears below $v \approx 0.85$. At $v \approx 0.9 - 2$, more or less coarsed lamellar morphology forms. When $c_0(x, y, 0) = \pm 0.1$, this limit where columnar morphology appears, shifts toward lower velocities.

The formation of oblique morphologies has also been observed. The most extensive OM-s appeared around the velocity $v \approx 1$, when oblique stripes with $c(x, y, 0) = 0.2$ and geometric parameters as above were initially introduced in the $x \in (5,30)$ space units region of the simulation area.

The most interesting characteristics of the pattern formation in a growing quenched area was found in the computational investigation of rotating fronts. Moving outwards along the radius, that is, reaching front segments with higher velocities, columnar morphology, oblique patterns forming different angles to the front, and, near the border of the quenched region, lamellar morphologies have been observed.
It is remarkable that the growing ends of the columnar stripes always remain perpendicular to the front (fig. 3). As a consequence, in the case of a front rotating with an appropriate speed, bent columnar structures build up, which develop into regular arcs. This result can be of major importance in various nanotechnological processes, since it makes possible the “wiring” of a surface upon a previously given curve. By moving the quenching front along an arbitrary non-intersecting path, a columnar structure of high and low density regions builds up behind it.

VII. CONCLUSION

The formation of striped microscopic patterns behind a traveling reaction-diffusion front in the  $NaOH + CuCl_2$  chemical system has been investigated. The scenario of the pattern formation was modeled by the terms of phase separation described by the Cahn-Hilliard Equation. The formation of the stripes, being parallel or oblique to the reaction-diffusion front, has been reproduced by computer simulations.

The results have been compared to the pattern formation in the case of directional quenching. At low front velocities, the formation of striped patterns perpendicular to the quenching front have been observed, these patterns being absent in the case of the source fronts. In the wake of a slowly progressing front that simultaneously changes its direction, the growing endpoints of the stripes will always be perpendicular to the front. This effect enables one to “draw” on a surface regular stripes following an arbitrary curve. Such a patterning could be of major importance in nanotechnology.

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[21] The spiral patterns in the classical two-dimensional Liesegang experiments [16] can also be considered as a special case of the oblique morphology.
[22] We used the nine-point Laplacian \( \Delta c(x_i, y_j) = \frac{1}{6}[4c(x_{i-1}, y_j) + 4c(x_{i+1}, y_j) + 4c(x_i, y_{j+1}) + 4c(x_i, y_{j-1}) + c(x_{i-1}, y_{j-1}) + c(x_{i+1}, y_{j-1}) + c(x_{i+1}, y_{j+1}) + c(x_{i+1}, y_{j+1}) - 20c(x_i, y_j)] \) and the fourth order term was approximated by \( \Delta^2 c = c(x_{i-2}, y_j) + c(x_{i+2}, y_j) + c(x_i, y_{j+2}) + c(x_i, y_{j-2}) + 2[c(x_{i-1}, y_{j-1}) + c(x_{i-1}, y_{j+1}) + c(x_{i+1}, y_{j-1}) + c(x_{i+1}, y_{j+1})] - 8[c(x_{i-1}, y_j) + c(x_{i+1}, y_j) + c(x_i, y_{j+1}) + c(x_i, y_{j-1})] + 20c(x_i, y_j) \)
FIG. 1: Stripes of colloidal CuO grains of oblique morphology. The precipitate structures form as a result of $8 \, M \, NaOH + 0.586 \, M \, CuCl_2$ reaction in a thin PVA gel sheet of about 0.2 $mm$ thickness, located between a microscope slide and a cover glass. The width of the figure is 0.58 $mm$. The sharp straight line shortly below the endpoints of the stripes represents the active border of the system.
FIG. 2: Pattern formation in the wake of source fronts traveling with constant speed. The dimensions of the simulation area are 250x620 space units. $\varepsilon = 0.5$, $a = 1$, $b = 10$, $\alpha = 0.1$ in all the sub-figures. A gray scale has been used, with white as $c = -1$ and black as $c = +1$. (a.) Random patterns formed far behind the front. Note the parallel striping that turns out in the wake of the random morphology. $v = 10$, $A = 1.78$ ($\varepsilon = 0$), $t = 50$. (b.) Regular lamellar patterns. $v = 1$, $A = 0.178$ ($\varepsilon = 0$), $t = 500$. (c.) Slightly disturbed lamellar morphology and oblique morphology with a small tilting angle formed when the first stripes were destroyed by random spots located in the $x \in (5, 30)$ space units region. $v = 1$, $A = 0.178$ ($\varepsilon = 0$), $t = 500$. (c.) Oblique morphology formed when the $x \in (5, 30)$ space units region was "pre-patterned" with tilted stripes forming an angle of about 30 degrees with the $Y$-axis and having a wavelength of about 8 space units. Note the parallel striping that turns out in the wake of the oblique morphology. $v = 1$, $A = 0.178$ ($\varepsilon = 0$), $t = 500$. 


FIG. 3: Pattern formation in the wake of rotating source fronts. The angular velocity is $\omega = 0.005$, the dimensions of the simulation area are 1200x600 space units. The parameter $\theta = 0.3$ was introduced to prevent the first stripes being parallel to one of the grid lines. $\varepsilon = 0.5$, $x_0 = 600$, $y_0 = 5$, $A = 0.00089$, $\alpha = 0.1$ ($c_f = 1$), $c_0 = -1$. Attention should be paid to the regions just behind the wake of the fronts; after this, coarsening will restructure the patterns. (a.) Oblique striping at $t = 100$. (b.) The critical angle is reached, the growth of the stripes becomes unstable. $t = 250$. (c.) Oblique morphology with a new angle builds up in the wake of the front. $t = 500$.14
FIG. 4: Pattern formation in the wake of a rotating quenching front. The dimensions of the simulation area are 600x1200 space units. The parameter $\theta_0 = 0.3$ was introduced to prevent the stripes being parallel to the grid lines. $\omega = 0.005$, $\epsilon = 0.5$, $x_0 = 600$, $y_0 = 5$, $c_0 = 0$; $t = 100$ (a.) and $t = 500$ (b.). Note the continuous arc-shaped stripes in the vicinity of the rotation center, where the local velocity is small. Attention should be paid to the regions just behind the front;