Eur. Phys. J. B **40**, 73–77 (2004)
DOI: 10.1140/epjb/e2004-00240-8

THE EUROPEAN PHYSICAL JOURNAL B

Transition to hexagonal pattern under the variation of intrinsic length scales of a reaction diffusion system

J. Sienkiewicz[1,a] and A. Bhattacharyay[2,b]

[1] Faculty of Physics, Warsaw University of Technology, ul. Koszykowa 75, 00-662 Warszawa, Poland
[2] Institut für Festkörperphysik, Technische Universität Darmstadt, Hochschulstr. 6, 64289 Darmstadt, Germany

Received 17 December 2003 / Received in final form 15 June 2004
Published online 3 August 2004 –

Abstract. The intrinsic length scales of a reaction diffusion system (Gierer-Meinhardt model) is varied by quasi-statically changing the diffusion constant of the activator and a transition from rolls to hexagon is detected. The transition is hysteretic or first order like. From stability analysis, we also analytically show the possibility of such transitions.

PACS. 87.10.+e General theory and mathematical aspects – 47.70.Fw Chemically reactive flows

Patterns like squares, hexagons, rhombus are observed in chemical systems [1], in Faraday surface wave ([2,3] and references therein), nonlinear optical systems [4] and in many other systems. Many reaction diffusion systems have been studied to show such structures under various ambient conditions. The origin, stability and dynamics of these patterns are subjects of active investigation [5–8]. In a reaction diffusion system the diffusivities are taken to be constants and the variation in them due to variation in temperature or other experimental conditions are considered to be on very small scales to produce any effect. We argue that if there are some sort of phase transitions which are hysteretic with the variation of diffusion constant, then it indeed has relevance to study transitions under the variation of diffusivities. In a situation when diffusion constants are very close to such transition points, a slight change in them due to the fluctuations in temperature etc. of the system might bring about a phase transition which will not be reversed when the diffusivities are restored. In another respect such studies are worth doing to understand how such a system behaves under a slow variation of its internal length scales. A slow variation of intrinsic length scale can be effected by slowly varying the diffusion constant of the system. In the present work we are going to present and characterize the phase transitions brought about by the variation of diffusivity of the activator of Gierer-Meinhardt (GM) [10] model keeping the diffusivity of the inhibitor fixed. In what follows, we will show roll patterns are loosing their stability to hexagonal structures via a first order like transition. Under the action of competing length scales, the patterns characterized by wave numbers which locally satisfy the condition $q_1+q_2+q_3+\ldots\ldots = 0$ are indeed an impressive general outcome and regular hexagons are a more symmetric special case of them.

The Gierer-Meinhardt model that we have taken up is

$$\frac{\partial A}{\partial t} = D_A \bigtriangledown^2 A + \rho_A \frac{A^2}{(1+K_A A^2)B} - \mu_A A + \sigma_A$$
$$\frac{\partial B}{\partial t} = D_B \bigtriangledown^2 B + \rho_B A^2 - \mu_B B + \sigma_B \qquad (1)$$

where A is the activator concentration and B is that of the inhibitor species. In what follows, we will always take the Turing [9] condition $D_B > D_A$ between the diffusivities of activator and inhibitor to hold true. In the above expression ρ_A and ρ_B are the reaction strengths, μ_A and μ_B are the self removal rates and σ_A and σ_B are the basic production terms for A and B respectively. In a parameter region where steady spatial order of concentration forms, the relevant length scale is given by [11]

$$\kappa^2 = \frac{C_1}{D_A} - \frac{C_2}{D_B}$$

where C_1 and C_2 are constants. Thus the diffusivities are the one which selects the internal length scale of the system. The present report comprises of three parts. The next part shows the results of numerical simulation done under quasi-static variation of the diffusion constant D_A. The following part accounts for the observations of numerical simulation on the basis of a stability analysis. The last part includes discussion.

In our simulation we have set $\sigma_A = \sigma_B = 0$. The other parameters are $\rho_A = \mu_A = 0.01$, $\rho_B = \mu_B = 0.02$, $K_A = 0.25$, $D_B = 0.2$ and D_A is varied from 0 to 0.01 by

[a] e-mail: julas@if.pw.edu.pl
[b] e-mail: arijit@fkp.tu-darmstadt.de

Fig. 1. The figure shows many domains of locally parallel rolls separated by domain walls and other defects at a value of the diffusivity $D_A = 0.005$. In this figure the concentration of the activator (A) has been plotted on a 256×256 lattice space.

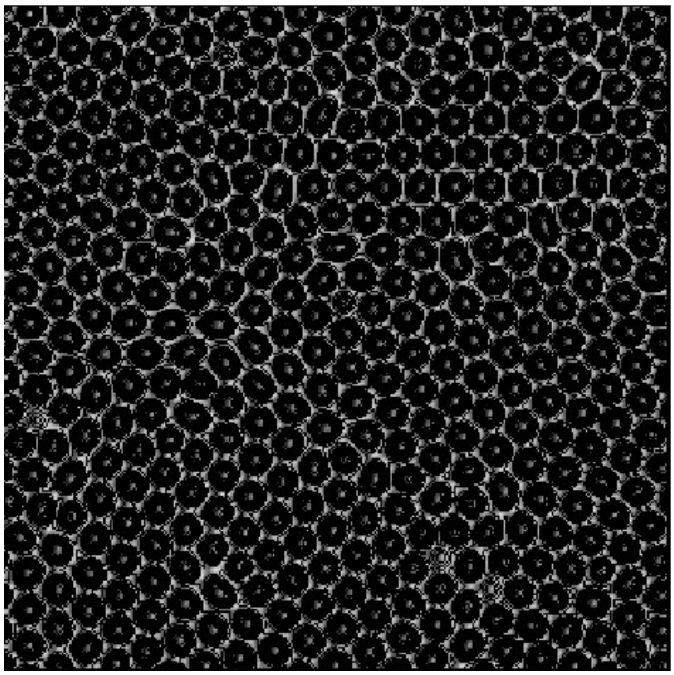

Fig. 2. The figure shows regular hexagonal order in the concentration of the activator species A at a value of the diffusivity $D_A = 0.0085$ on the same space lattice as Figure 1.

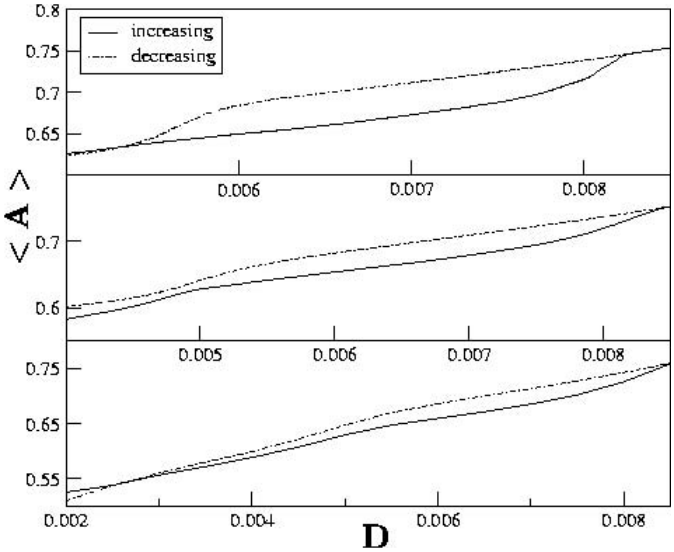

Fig. 3. The figure shows a plot for the hysteresis loops obtained at the transition from rolls to hexagons and back for the rate of variation of D_A 0.00001, 0.0001 and 0.0005 from top to bottom. The broken upper curves are paths along which the average conc. of A varies when D_A is decreasing.

the steps of 10^{-5} near the transition regions and by the steps of 10^{-4} in other parts keeping 10^4 temporal steps between every variation of D_A. The simulation is done on two dimensions by implementing finite difference method and on a lattices of size 256×256. In the first part of our two dimensional simulation so long as D_A remains less than approx. 0.004, the locally isolated concentration peaks show up. Such structures appear for very small value of D_A compared to D_B because of the fact that the activator finds no time to spread compared to very fast inhibitor spreading. As D_A is increased, near $D_A = 0.004$ steady roll structures which are locally parallel appear and persist up to $D_A < 0.008$. As Figure 1 shows, many small domains of parallel rolls are present and are oriented in all possible directions.These domains are separated by very many defects and domain walls. At about $D_A = 0.008$ a transition from rolls to almost regular looking hexagonal structures takes place. This phase is stable up to a D_A value about 0.009. On a further increase in D_A, hexagonal patterns loose its stability to a steady homogeneous state. Figures 1 and 2 show the roll structure at $D_A = 0.005$ while increasing D_A and hexagonal at $D_A = 0.0085$ as seen in the lattice. After the hexagons have stabilized at $D_A > 0.008$, now if we go on decreasing D_A the rolls do not come back at $D_A = 0.008$ but at a much lower value. The transition from rolls to hexagons show hysteresis. A plot of average concentration of A against D_A is shown in Figure 3 (topmost part). This plot clearly shows a jump in the average concentration as we go from below along D_A axis which marks the transition to hexagons from rolls at about $D_A = 0.008$. As we come down the D_A axis, we see that the average amplitude does not immediately jump back to the lower line at $D_A = 0.008$ but continues to follow the same upper line representing the hexagons and comes back to the lower value at around $D_A = 0.0055$ which marks transition to rolls from hexagons. Thus, Figure 3 shows hysteresis and identifies the transition to be first order like. The average activator amplitude also comes out as a relevant parameter to characterize such situations. The

graphs in the middle and the lowest part in Figure 3 are the same plots but for the rate of change of D_A at every time step by the amount 0.0001 and 0.0005 respectively. It is clear from the lower two parts of the Figure 3 that the loop spreads out with the comparatively rapid variation of D_A and the upper (decreasing D_A) and the lower (increasing D_A) lines are coming more close making the hysteresis loops less clearly defined. It appears that the phase transition does not remain a well defined first order like transition at all the time scales of variation of the parameter D_A and tend to become more and more continuous at faster variations of D_A. Such a dependence of the nature of transition on the dynamics of variation of the parameter is very important.

Having numerically found the results let us try to explain it from analytics. After rescaling of concentration, length and time as $A = \mu_A A$, $l = \sqrt{\frac{D_B}{\mu_A}} l$ and $t = \mu_A t$ respectively, equation (1) takes the form

$$\frac{\partial A}{\partial t} = \bar{D}\bigtriangledown^2 A + \bar{\rho_A}\frac{A^2}{(1+\bar{K}A^2)B} - A$$
$$\frac{\partial B}{\partial t} = \bigtriangledown^2 B + \bar{\rho_B}A^2 - \bar{\mu_B}B \qquad (2)$$

where we have taken $\sigma_A = \sigma_B = 0$. Under such scaling we have $\bar{D} = \frac{D_A}{D_B} < 1$, $\bar{\rho_A} = \frac{\rho_A}{\mu_A^2}$, $\bar{K} = \frac{K_A}{\mu_A^2}$, $\bar{\rho_B} = \frac{\rho_B}{\mu_A^3}$, and $\bar{\mu_B} = \frac{\mu_B}{\mu_A}$. The homogeneous steady fixed point of the system is $A = \overset{*}{A} = 0.0085$ (under prevalent conditions of the simulation) and $B = \overset{*}{B} = \frac{\bar{\rho}_B \overset{*}{A}^2}{\bar{\mu}_B} = 0.72$. A linear stability analysis shows that inhomogeneous perturbations will grow with a growth rate λ given by

$$\lambda = -\frac{1}{2}\left[\bar{D}k^2 + \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B} - 1 + k^2 + \bar{\mu}_B\right] \pm \left[\left\{\left(1 - \bar{D}k^2 - \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B}\right) + (k^2 + \bar{\mu}_B)\right\}^2 - 8\bar{\mu}_B\right]^{1/2}. \qquad (3)$$

In the range of given parameter values the real part is always negative giving no scope for Hopf modes to appear and in our simulation we have not seen any oscillatory phase. Since the real part of the growth rate is negative, an inhomogeneous perturbation will only grow when

$$\left[1 - \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B} - (\bar{D}+1)k^2 - \bar{\mu}_B\right]^2 \leq \left[1 - \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B} + (1-\bar{D})k^2 + \bar{\mu}_B\right]^2 - 8\bar{\mu}_B. \qquad (4)$$

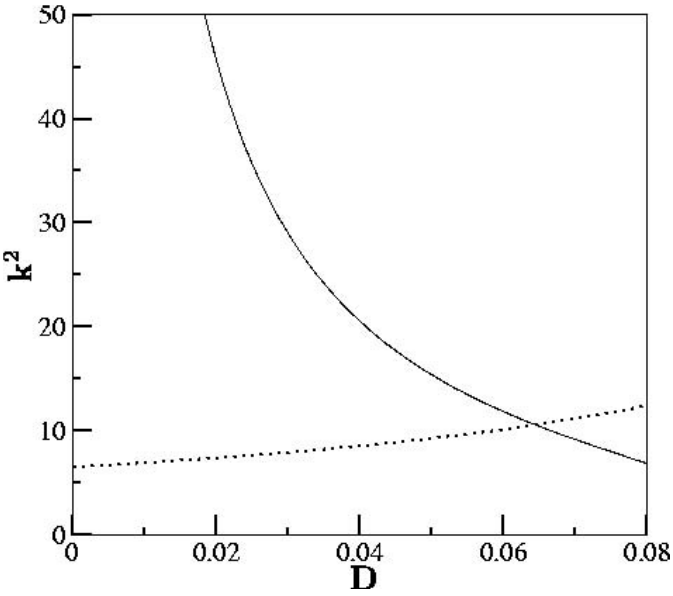

Fig. 4. The figure shows the phase diagram as obtained from linear stability on a $D - k^2$ plane. The roll pattern is stable in the region below the continuous curve. The hexagons are stable anywhere above the dotted curve.

Now, the left hand side of the above inequality is always positive. thus the above condition is also the condition for the growth rate to be real. After simplifying the above inequality we get the relevant wave number satisfying the relation

$$k^4 + k^2\frac{D\bar{\mu}_B - \left(1 - \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B}\right)}{D} + \frac{\bar{\mu}_B\left(1 + \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B}\right)}{D} \leq 0. \qquad (5)$$

Given the above condition valid we get a range for the allowed k value given by $k_1^2 < k^2 < k_2^2$, where k_1 and k_2 reads

$$k_{1,2}^2 = \frac{1 - \left(D\bar{\mu}_B + \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B}\right)}{2D} \pm \frac{\sqrt{\left(D\bar{\mu}_B + 1 - \frac{2\bar{K}\overset{*}{A}^3\bar{\rho}_B}{\bar{\rho}_A\bar{\mu}_B}\right)^2 - 8D\bar{\mu}_B}}{2D}. \qquad (6)$$

In the given parameter range k_1^2 is negative, so $0 < k^2 < k_2^2$ and the boundary is shown in Figure 4 by the continuous line below which the rolls are stable in the $D - k^2$ plane. We see that, k^2 varying as $1/D$, the growth rate λ approaches zero as we go on increasing D. The growth rate decreases because the discriminant in equation (3), which contributes positively to the growth rate λ, decreases with increasing D. Now, by increasing D_A we are actually increasing D in our simulation and thus approaching the upper instability boundary where the growth rate of the rolls are small. In such a region other slow modes become comparable to the pre-existing rolls and an interaction in them is responsible for the creation of hexagonal pattern.

Let us do simple calculations in order to see the possibility of a hexagonal instability to develop near the upper instability boundary of the rolls. Interesting things show up if we try to see how a perturbation grows which tends to rotate the existing roll solutions. To do so we perturb the roll solutions as $A(1+\delta a\cos(\mathbf{k_1}\cdot\mathbf{r}))\cos(\mathbf{k}\cdot\mathbf{r})$ so as to probe a possible rotation of the locally parallel rolls and we end up with two sets of linear decoupled equations in δa and δb obtained from harmonic balance of the terms containing $\cos(\mathbf{k_1}+\mathbf{k})\cdot\mathbf{r}$ and $\cos(\mathbf{k_1}-\mathbf{k})\cdot\mathbf{r}$. The equations are

$$\begin{aligned}\left(\bar{K}+\frac{3A^2}{4}\right)\frac{\partial\delta a}{\partial t} &= -\left[\bar{D}\left(\bar{K}+\frac{A^2}{2}\right)|\mathbf{k_1}+\mathbf{k}|^2\right.\\ &\quad\left.+\bar{D}|\mathbf{k_1}-\mathbf{k}|^2\frac{A^2}{4}\right]\delta a+\frac{\bar{\rho}_A\bar{K}A}{B}\delta a\\ &\quad-\left(\bar{K}+\frac{3A^2}{2}\right)\delta a\\ \frac{\partial\delta b}{\partial t} &= -|\mathbf{k_1}+\mathbf{k}|^2\delta b-\bar{\mu_B}\delta b \qquad (7)\end{aligned}$$

and

$$\begin{aligned}\left(\bar{K}+\frac{3A^2}{4}\right)\frac{\partial\delta a}{\partial t} &= -\left[\bar{D}\left(\bar{K}+\frac{A^2}{2}\right)|\mathbf{k_1}-\mathbf{k}|^2\right.\\ &\quad\left.+\bar{D}|\mathbf{k_1}+\mathbf{k}|^2\frac{A^2}{4}\right]\delta a+\frac{\bar{\rho}_A\bar{K}A}{B}\delta a\\ &\quad-\left(\bar{K}+\frac{3A^2}{2}\right)\delta a\\ \frac{\partial\delta b}{\partial t} &= -|\mathbf{k_1}-\mathbf{k}|^2\delta b-\bar{\mu_B}\delta b. \qquad (8)\end{aligned}$$

It is apparent from the shape of above two sets of equations that a simultaneous validity of them requires $\mathbf{k_1}$ to have two values such as $\mathbf{k_1}=\mathbf{k_{11}}$ and $\mathbf{k_1}=\mathbf{k_{12}}$ where $|\mathbf{k_{11}}|=|\mathbf{k_{12}}|$ and $\mathbf{k_{11}}$ & $\mathbf{k_{12}}$ are at angles θ and $\pi\pm\theta$ with the direction of $\mathbf{k}$. Such a choice of k_1 predicts development of rhombic structures at the cost of rolls. But we have not been able to stabilize such structures. A regular hexagon grows when $|\mathbf{k_{11}}|=|\mathbf{k_{12}}|=|\mathbf{k}|$ and $\theta=60^\circ$ along with the case $|\mathbf{k_{11}}|=|\mathbf{k_{12}}|=0$ where the original roll grows on the same footing as the combined ones to form the hexagon. In such a situation we get all the three wave numbers, one being that of the preexisting roll whose growth rate is the same as other two new modes which are locally rotated at 60° with it, grow equally in the concentration of the activator A obeying the equation as follows

$$\begin{aligned}\left(\bar{K}+\frac{3A^2}{4}\right)\frac{\partial\delta a}{\partial t} &= -\bar{D}|\mathbf{k}|\left(\bar{K}+\frac{3A^2}{4}\right)\delta a+\frac{\bar{\rho}_A\bar{K}A}{B}\delta a\\ &\quad-\left(\bar{K}+\frac{3A^2}{2}\right)\delta a.\end{aligned}$$

Here A is the amplitude of the steady roll pattern and we will treat it as a constant such that $\bar{K}A^2$ is of $O(1)$. We will do so without trying to actually solve for A because it takes to solve an equation which is 7th order in the power of A to get it as a fixed point of the system at least in the presence of a homogeneous steady state. The above approximation is quite reasonable in view of the fact that $\bar{K}$ is the saturation constant of the system. Moreover the basic homogeneous steady state has a very small concentration in the activator which implies the saturation is achieved mainly by the contribution from the steady roll amplitude. The ratio A/B can easily be shown to be equal to $(k^2+\bar{\mu}_B)/2a_0\bar{\rho}_B)$ where a_0 is the concentration of the activator of the homogeneous state in presence of which a steady roll develops. In what follows we will take a_0 of the same order as the basic homogeneous steady state. With all these things taken into account the growth rate is given by

$$\begin{aligned}\lambda_{hex} &= \left(\frac{\bar{\rho}_A}{2a_0\bar{\rho}_B}-D\left[1+\frac{3}{4}\bar{K}A^2\right]\right)k^2\\ &\quad+\left(\frac{\bar{\rho}_A\bar{\mu}_B}{2a_0\bar{\rho}_B}-\left[1+\frac{3}{2}\bar{K}A^2\right]\right). \qquad (9)\end{aligned}$$

The second part of the above expression is negative. Thus, the growth rate is positive for

$$D\le D_{max}=\frac{2\bar{\rho}_A}{7a_0\bar{\rho}_B}. \qquad (10)$$

So we approximately get $D\le 0.17$ which sets the upper boundary for the hexagonal phase to exist. Within this range of diffusion constant the wave number for the hexagonal pattern on the instability boundary is given as

$$k^2\ge\frac{\frac{5}{2}-\frac{\bar{\rho}_A\bar{\mu}_B}{2a_0\bar{\rho}_B}}{\frac{\bar{\rho}_A}{2a_0\bar{\rho}_B}-\frac{7D}{4}}. \qquad (11)$$

This curve is also plotted in Figure 4 as a broken line. The region above this line (and for $D\le D_{max}$) is where a hexagonal instability grows. Since the typical wave number k of hexagon goes as $\sqrt{C1/(C2-D)}$, where $C1$ and $C2$ are constants, its the least at $D=0$ and the growth rate being proportional to k^2 is small and cannot compete with rolls which has its typical wave number as $\sqrt{1/D}$ and higher growth with bigger k. Suppose the role state formed at small D persists up to its upper instability boundary. Let us look at Figure 5 which is the same as Figure 4 along with some extra diagram. When one crosses the upper instability boundary for rolls at the point **A** towards hexagons by increasing D, a transition from rolls to hexagons takes place. Let us take $D=D_{forward}$ where the transition occurs. A subsequent increase in D might result in an increase of the wave number of the hexagon because the typical wave number goes as $\sqrt{C1/(C2-D)}$. Since growth rate of hexagons are higher for higher k the wave number can also grow to some extent even at a fixed D. An increase in the wave number takes us at a higher point in the $D-k^2$ plane than where we actually crossed the instability boundary. Now, if we start decreasing D and go backward the path along which we move should be that horizontally crossing the instability boundary at **B** in Figure 5. This is so because the

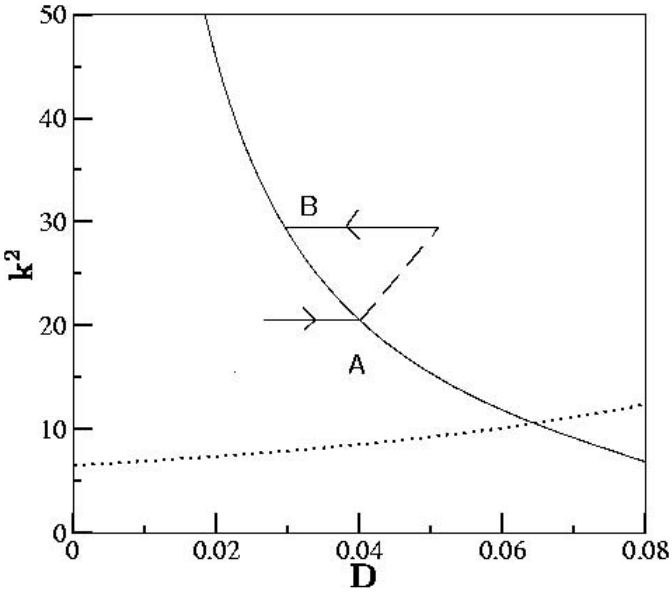

Fig. 5. This figure shows the same phase diagram as in Figure 4. It has been considered that the forward and backward transitions from rolls to hexagons are taking place at points **A** and **B** respectively

decrease in D should not result in an associated decrease in the wave number k to slower the growth. Now, the backward transition taking place at $D = D_{backward}$, the region of hysteresis is given by $D_{hyst} = D_{foward} - D_{backward}$. A comparatively rapid variation of D near the instability boundary may not simply allow the system in the hexagonal phase to attain correspondingly higher wave numbers and thus while coming back it will lower the point **B** where the backward transition takes place. So, the region of overlap will decrease and thus, from simple arguments, we get a qualitative information about the dependence of the hysteresis on the rate of the variation of the diffusivity.

In the conclusion, we would like to mention that a transition from rolls to hexagons has been observed with increasing diffusivity of the activator species in the GM model. The transition shows well defined region of hysteresis when the diffusivity is varied on a very slow rate. With the increase in the variation rate of D the region of hysteresis becomes shorter. We also qualitatively show from simple analytic arguments that there can be a first order like phase transition from rolls to hexagons. We have numerically seen that the hexagons loose stability to a homogeneous steady state at higher diffusivity. Calculations also show that there is an upper D value marked as D_{max} beyond which hexagons would not grow. The fact that rolls are preferred to hexagons at smaller D are qualitatively accounted for from the knowledge of the typical wave numbers and growth rates of the respective phases. Let us make a comment on the decoupled sets of equations – equation (7) and equation (8) – which we have got because we have not taken into account the homogeneous steady state in presence of which a nonlinear roll solution can stabilize. Had we taken that state into consideration when we perturbed the rolls we would have got coupled equations. Since the presence of the homogeneous steady state does not play a role in the formation of hexagonal instability, which is generated by the nonlinear interaction of the instability ($\cos(\mathbf{k_1} \pm \mathbf{k}) \cdot \mathbf{r}$ terms) with the preexisting rolls, and moreover considering the very small value of the activator concentration in the homogeneous steady state we think that it would have negligible effect to alter the qualitative results arrived at except increasing complexity of the algebra. Thus, although the inhibitor parts in the decoupled equations actually show decay one can imagine that if the coupling is restored it will also grow to produce the corresponding structure to the activator.

J.S. would like to express his gratitude to the organizers of the "German-Polish-Lithunian Dialogue 2003" (Especially the hosting group of B. Drossel). He also acknowledges the financial support of the Warsaw University of Technology.

References

1. Q. Ouyang, H.L. Swinney, Nature (London) **352**, 610 (1991)
2. C. Wagner, H.W. Müller, K. Knorr, Phys. Rev. E **62**, R33 (2000)
3. H. Arbell, J. Feinberg, Phys. Rev. E **65**, 036224 (2002)
4. R. Neubecker, O. Jacoby, Phys. Rev. E **67**, 066211 (2003)
5. G.H. Gunaratne, Q. Ouyang, H.L. Swinney, Phys. Rev. E **50**, 2802 (1994)
6. B. Echebarria, H. Riecke, Phys. Rev. Lett. **84**, 4838 (2000)
7. B. Pena, C. Perez-Garcia, Phys. Rev. E **64**, 056213 (2001)
8. A.A. Golovin, A.A. Nepomnyashchy, Phys. Rev. E **67**, 056202 (2003)
9. A.M. Turing, Philos. Trans. R. Soc. London, Ser. B **237**, 37 (1952)
10. A. Gierer, H. Meinhardt, Kybernetik **12**, 30 (1972)
11. A.J. Koch, H. Meinhardt, Rev. Mod. Phys. **66**, 1481 (1994)