Periodic spatiotemporal patterns in a two-dimensional two-variable reaction-diffusion model

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Periodic spatiotemporal two-dimensional (2D) asymptotic patterns in an excitable two-variable thermochemical (reaction-diffusion) system are shown. In a one-dimensional system the traveling impulse which reflects from impermeable boundaries is a stable asymptotic solution if the diffusion coefficient of the reactant is greater than the thermal diffusivity of the system. Periodic patterns of two symmetries are presented in the 2D system: the impulse of excitation propagating along the diagonal of a square spatial domain and a structure consisting of curved impulses which propagate in the direction perpendicular to one side of a rectangular domain.

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I. INTRODUCTION

Reflections of traveling impulses, their splitting [1,2], and spatiotemporal chaos [3] have been reported in two-variable one-dimensional (1D) reaction-diffusion (RD) activator-inhibitor systems for the diffusion coefficient of the inhibitor greater than the diffusion coefficient of the activator. The reflection of the impulse from impermeable boundaries has been reported in two-variable 1D RD models such as the Gray-Scott model [4], a model describing an enzymatic reaction inhibited by an excess of its reagents [5], and a model of an exothermic reaction for which the rate constant obeys the Arrhenius formula [6]. The reflection of the traveling impulse has also been observed in the four-variable Oregonator model close to the subcritical Hopf bifurcation, where limit cycle oscillations coexist with stable focus [7]. The reflection of chemical wave packets has been reported in the two-dimensional (2D) three-variable Brusselator model [8,9]. Similar results have been obtained in the modified Oregonator model which has been used by authors for the qualitative description of the nonlinear phenomena observed in the Belousov-Zhabotinsky reaction in a dispersed oil-water system [10]. The refraction and reflection of the front wave have been observed experimentally in the Belousov-Zhabotinsky reaction on a line of discontinuity separating two parts of the system in which the chemical kinetics is different [11]. These phenomena have been also studied using the kinematic approach based on the eikonal approximation [12].

In the present paper the possibility of existence of stable asymptotic 2D periodic spatiotemporal solutions in a two-variable excitable activator-depleted reactant thermochemical model is considered. Temperature corresponds to the activator and the depleted reactant plays the role of the inhibitor in commonly used activator-inhibitor systems. If the diffusion coefficient of the reactant is sufficiently greater than the thermal diffusivity of the reacting mixture, then the stable asymptotic solution in a 1D bounded system has the form of a traveling impulse which reflects from the boundaries and spreads periodically from one wall to the other. This condition corresponds to a low Lewis number ($\mathrm{Le}<1$). Such conditions are not met in liquid systems but they can be satisfied in the gaseous phase. Examples of systems with $\mathrm{Le}<1$ are H_2-O_2-N_2 mixtures in which instability of flame fronts has been observed [13]. In a square spatial domain and for symmetric initial excitations, we show a 2D impulse traveling along the diagonal and periodically reflecting from the boundaries. Such a symmetric impulse is stable only for a spatial domain whose size is smaller than some critical value. Moreover, we present a periodic spatiotemporal structure which is the result of instability of a planar impulse in a 2D system. The structure consists of two curved traveling impulses reflecting from the boundaries as well as from each other. The curved impulses propagate parallel to the longer sides of the rectangle. The instability of the planar and circular impulses has been shown by numerical calculations in the same model as that considered in the present paper [14]. The authors have shown the breaking of impulses only.

In the next section we present a model of a 2D two-variable reaction–diffusion–thermal-conduction system. The main results are presented in the subsequent section, and the conclusions are presented in the last section.

II. MODEL

The model describes an open thermochemical system in which one irreversible exothermic reaction occurs:

$$S \xrightarrow{k} P + Q. \tag{1}$$

The rate constant of the reaction depends on temperature T according to the Arrhenius formula. We consider the thin cuboidal $(x,y,z) \in [0,L_x]\times[0,L_y]\times[0,h]$ system in which the reaction (1) occurs. The vertical boundaries of the system are thermally isolated. They are also impermeable for the reactant S. The heat and the reactant are exchanged with the reservoir placed at $z=0$. The temperature and the concentration of the reactant in the reservoir are equal to T_0 and S_0, respectively. The heat transfer with the reservoir obeys the Newton law with the heat transfer coefficient α. The exchange of the reactant between the reservoir and the system

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is described by $k_0(S_0-S)$. The reaction heat Q, the heat transfer coefficient α, the heat capacity of the reacting mixture C_p, the density of the reacting mixture ρ, and the thermal conductivity of the reacting mixture λ are assumed to be constant. We assume that the height of the system h is so small that the changes of the temperature and the reactant concentration along the z coordinate may be neglected. Therefore, the space-time behavior of the system is described by the following equations:

$$\frac{\partial S}{\partial t'} = D_S\left(\frac{\partial^2 S}{\partial x'^2} + \frac{\partial^2 S}{\partial y'^2}\right) + k_0(S_0 - S) - kS, \tag{2a}$$

$$\frac{\partial T}{\partial t'} = D_T\left(\frac{\partial^2 T}{\partial x'^2} + \frac{\partial^2 T}{\partial y'^2}\right) + \frac{Q}{C_p\rho}kS - \alpha(T - T_0), \tag{2b}$$

where D_T is the thermal diffusivity of the reacting mixture $D_T=\lambda/(C_p\rho)$ and D_S is the diffusion coefficient of the reactant. Taking into account the Arrhenius formula for the rate constant k, the dynamics of the system in dimensionless variables may be described by the following equations [15]:

$$\frac{\partial s(x,y,t)}{\partial t} = D\left(\frac{\partial^2 s}{\partial x^2} + \frac{\partial^2 s}{\partial y^2}\right) + A_1(s_0 - s) - e^{\theta/(1+A_2\theta)}s, \tag{3a}$$

$$\frac{\partial \theta(x,y,t)}{\partial t} = \frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} + B(e^{\theta/(1+A_2\theta)}s - \theta), \tag{3b}$$

where $x=\sqrt{k(T_0)/D_T}x'$ and $y=\sqrt{k(T_0)/D_T}y'$ are dimensionless spatial coordinates, $t=k(T_0)t'$ is the dimensionless time, $\theta=E(T-T_0)/(RT_0^2)$ is the dimensionless temperature, $s=S/\{C_p\rho RT_0^2\alpha/[k(T_0)QE]\}$ is the dimensionless concentration of the reactant, $A_1=k_0/k(T_0)$, $s_0=S_0/\{C_p\rho RT_0^2\alpha/[k(T_0)QE]\}$, $A_2=RT_0/E$, $B=\alpha/k(T_0)$, and $D=D_S/D_T$ are dimensionless parameters, E is the activation energy for the reaction, and R is the gas constant.

In order to define the problems we assume the initial conditions for a square domain:

$$s(x,y,0) = s^*, \quad \theta(x,y,0) = \theta^* \text{ for } (x,y) \in [0,l_x^*] \times [0,l_y^*],$$

$$s(x,y,0) = s_\infty, \quad \theta(x,y,0) = \theta_\infty \text{ for other points}, \tag{4a}$$

where l_x^* and l_y^* denote the sizes of the region of the initial excitation.

For a rectangular domain the initial distributions have the form

$$s(x,y,0) = s(x,t_0), \quad \theta(x,y,0) = \theta(x,t_0) + \text{pert}(x,y),$$

$$(x,y) \in [0,L_x] \times [0,L_y] \tag{4b}$$

where $s(x,t_0)$ and $\theta(x,t_0)$ are profiles of the planar impulse at $t=t_0$ and pert indicates the perturbation.

The boundary conditions have the following form:

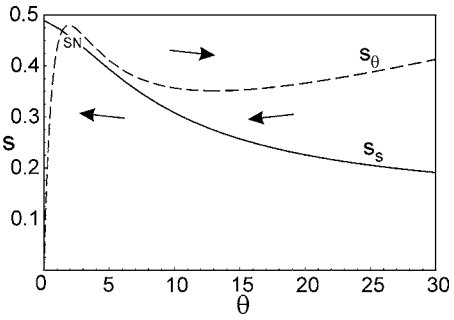

FIG. 1. Nullclines for the activator (s_θ) and the inhibitor (s_S) for Eqs. (3) without the diffusion terms for $A_1=45$, $s_0=1/2$, and $A_2=1/5$. Arrows schematically show directions of the vector field.

$$\frac{\partial s(0,y,t)}{\partial x} = \frac{\partial s(L_x,y,t)}{\partial x} = 0, \quad y \in [0,L_y],$$

$$\frac{\partial s(x,0,t)}{\partial y} = \frac{\partial s(x,L_y,t)}{\partial y} = 0, \quad x \in [0,L_x];$$

$$\frac{\partial \theta(0,y,t)}{\partial x} = \frac{\partial \theta(L_x,y,t)}{\partial x} = 0, \quad y \in [0,L_y];$$

$$\frac{\partial \theta(x,0,t)}{\partial y} = \frac{\partial \theta(x,L_y,t)}{\partial y} = 0, \quad x \in [0,L_x]. \tag{5}$$

We assume the following values of the parameters: $A_1=45$, $s_0=1/2$, and $A_2=1/5$. At these values of the parameters, the system [Eqs. (3)] without the diffusion terms is excitable and has one stationary state, namely, the stable node in which the temperature and the concentration of the reactant are $\theta_\infty=1.399\,262\,529$ and $s_\infty=0.468\,905\,277$, respectively (see Fig. 1). The values of the excitation $\theta^*=10$ and $s^*=0.44$ belong to the area between the middle and the right-hand branch of the nullcline for the temperature.

III. RESULTS

It is noteworthy that our system is excitable and far from any bifurcation because the single stationary state is the stable node. The temperature is the activator and the depleted reactant plays the role of inhibitor. It should be stressed that the decrease of the stationary value of s makes the system less excitable (see Fig. 1). The analytical solutions to Eqs. (3)–(5) are unknown and therefore Eqs. (3)–(5) are solved numerically. We use the Cranck-Nicholson scheme with the spatial steps equal to 2×10^{-2} for diffusion terms, and the fourth-order Runge-Kutta algorithm with the time step equal to 4×10^{-6} for kinetics terms. The asymptotic solution to Eqs. (3)–(5) in a 1D infinite system has the form of a traveling impulse $s(\xi)$ and $\theta(\xi)$ where $\xi=x-ct$ and c is its velocity. In a finite 1D system the traveling impulse reflects from the boundaries and spreads periodically from one wall to the other. An example of the reflection of the traveling impulse is shown in Fig. 2(a). After initial excitation the impulse propagates right as is shown in Fig. 2(a). Let us

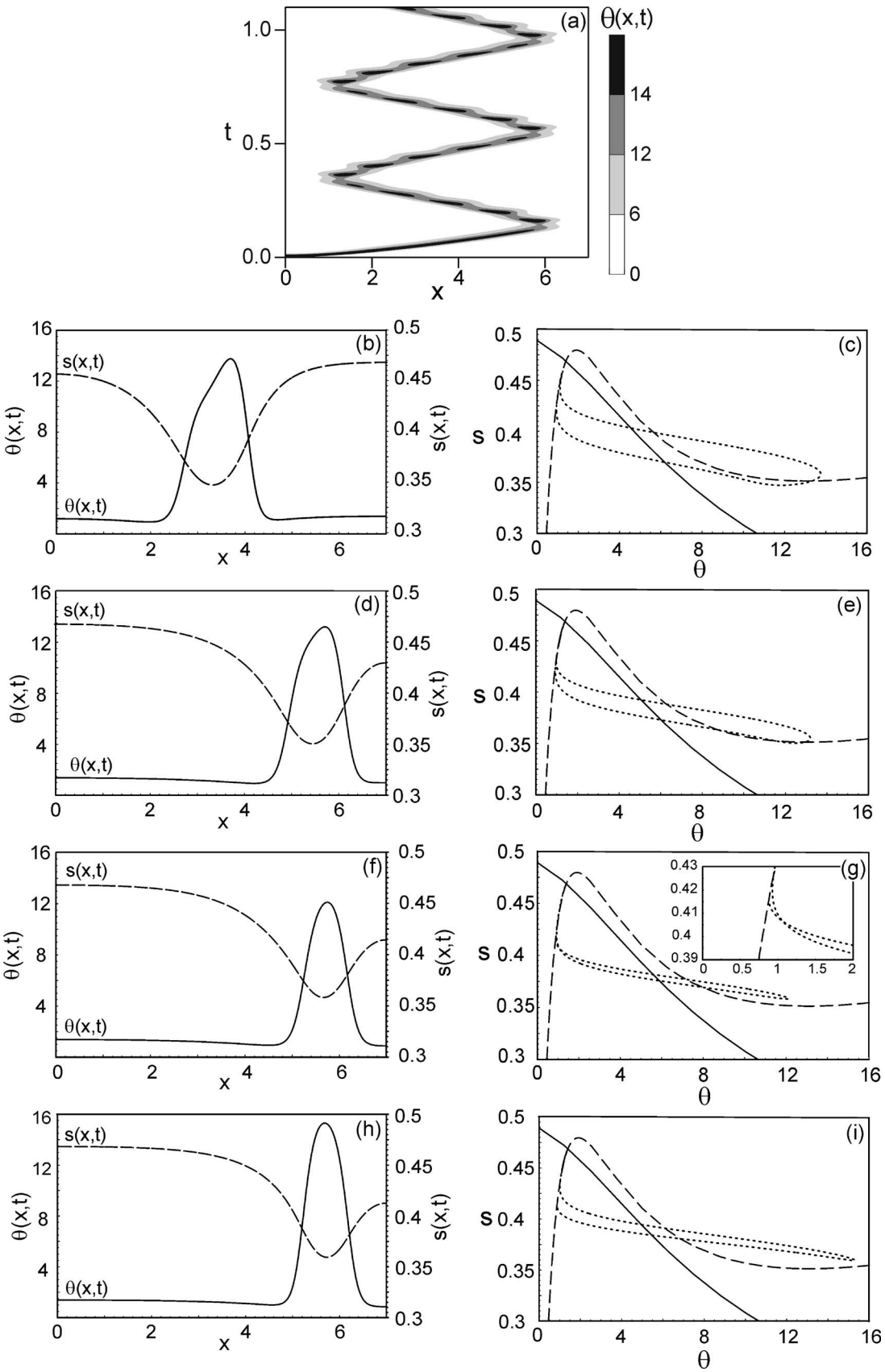

FIG. 2. 1D solution to Eqs. (3), (4a), and (5) in the form of a reflecting impulse for $B=2000$ and $D=3.5$ (a). The remaining parameter values are the same as in Fig. 1. The profiles of temperature $\theta(x,t)$, the concentration of the reactant $s(x,t)$, and projections of the solutions on the plane (θ,s) for $t=0.458$ (b), (c), 0.538 (d), (e), 0.550 (f), (g), and 0.568 (h), (i) are shown below.

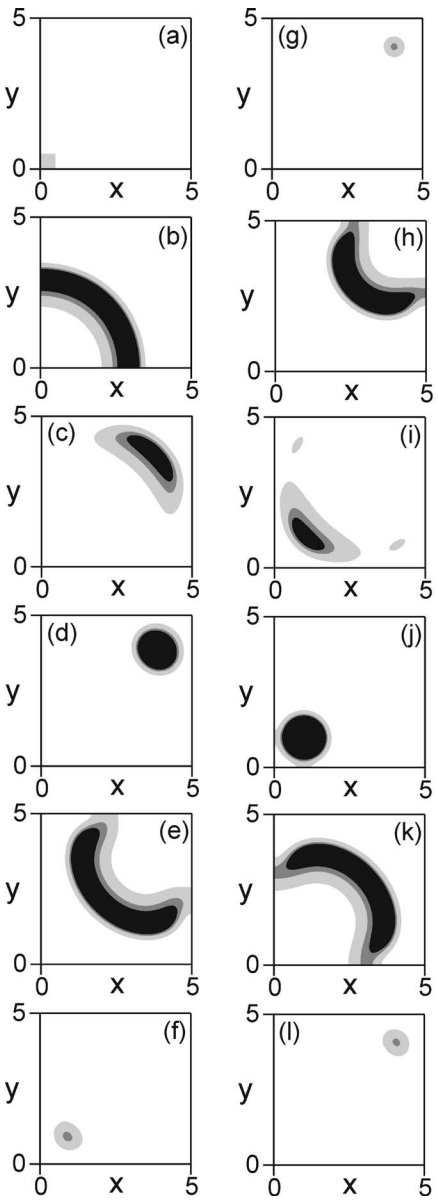

FIG. 3. Solutions to Eqs. (3), (4a), and (5) for $l_x^*=l_y^*=0.5$, $\theta=10$, and $s^*=0.44$ at $t=0$ (a), 0.04 (b), 0.08 (c), 0.12 (d), 0.16 (e), 0.2 (f), 1.56 (g), 1.6 (h), 1.64 (i), 1.68 (j), 1.72 (k), and 1.76 (l). The remaining parameter values are the same as in Fig. 2.

mention that just after reflection the shape of the impulse is not invariable but changes periodically. However, these oscillations decay with the increase of the size of 1D system. In Fig. 2(c) the projection of the distributions of θ and s presented in Fig. 2(b) is shown on the plane (θ,s). The upper (lower) part of the dotted curve on Fig. 2(c) corresponds to the front (back) of the traveling impulse. The propagation of the impulse is controlled by the threshold for excitation which is seen in Figs. 2(c), 2(e), 2(g), and 2(i) as the interval of the front projection between the point with the values of θ and s at the right boundary and the point of the intersection with the nullcline for θ. The velocity of the impulse is determined by the threshold because the part of the trajectory below the threshold is situated in the region of the phase plane where the vector field is directed to the stationary state. When the impulse approaches the right boundary the value of s at the boundary decreases [compare Figs. 2(b) and 2(d)], which causes the increase of the threshold, because the intersection point of the front projection is shifted to lower values of s and larger values of θ [compare Figs. 2(c) and 2(e)]. Therefore, the velocity of the impulse decreases and the front of the impulse stops. Further decrease of s at the boundary causes the intersection of the parts of the projection corresponding to the front and the back [see Fig. 2(g) and inset there]. Next the exchange of the front with the back occurs in such a way that the former front becomes the current back and the former back forms the current front [see Fig. 2(i)]. The impulse reflects from the boundary and starts moving in the opposite direction [see Fig. 2(h)]. The same scenario repeats when the impulse approaches the left boundary. The impulse reflects provided that the value of D is greater than the critical value D_{cr}. For the values of parameters used in this paper and $B=2000$, D_{cr} is approximately equal to 3.28. For D less than D_{cr} the impulse disappears at the boundary because of the zero-flux boundary conditions. The critical value D_{cr} increases when the value of B increases. Moreover, the impulse reflects provided that the length of the system L is greater than the critical value L_{cr}. For shorter lengths the excitation disappears at the right boundary, because the distribution of θ with the amplitude situated below the nullcline for θ is created when the impulse reaches the right boundary. The initial excitation does not approach a form sufficiently close to the asymptotic traveling impulse because the system is too small. For the values of the parameters used in this paper, $B=2000$ and $D=3.5$, L_{cr} is approximately equal to 5.40. Because of the equivalence of the reflection from the impermeable boundary and the reflection of impulses from each other, the result of a collision of two impulses depends on the distance between the places of their generation. Let us mention that Eqs. (3) are invariant to the change x to $(-x)$, which follows in the mirror symmetry of their solutions. The solution to Eqs. (3) on the interval $[0,L]$ and its mirror reflection on the interval $[L,2L]$ form the solution on the interval $[0,2L]$. Therefore, the reflection of the traveling impulse from the impermeable boundary is equivalent to the reflection of two identical impulses. It should be stressed that for $B=2000$ the 1D traveling impulse, which reflects from the boundaries, is the structurally stable asymptotic solution for $D\in[3.5,5.75]$. For values of $D>5.75$ the backfiring phenomenon [16] is observed.

In 2D square domains ($L_x=L_y=L$) and for symmetric initial conditions ($l_x^*=l_y^*$) [see Fig. 3(a)] a traveling impulse, symmetric with respect to the diagonal, is formed. The impulse reflects from the corner along the diagonal [Figs. 3(c)–3(e)] provided that L is sufficiently large. If L is shorter, then the impulse disappears at the boundary. After some transient period the impulse reflects periodically from the corners [Figs. 3(b)–3(l)] and the period of the system approaches its asymptotic value. One can see that the system in Figs. 3(g) and 3(l) is nearly in the same phase. The stability of such an asymptotic solution was checked by small perturbations at various points of the numerical grid. The periodically reflecting impulse is stable provided that L is less than the critical value L'_{cr}. For the values of the parameters used in

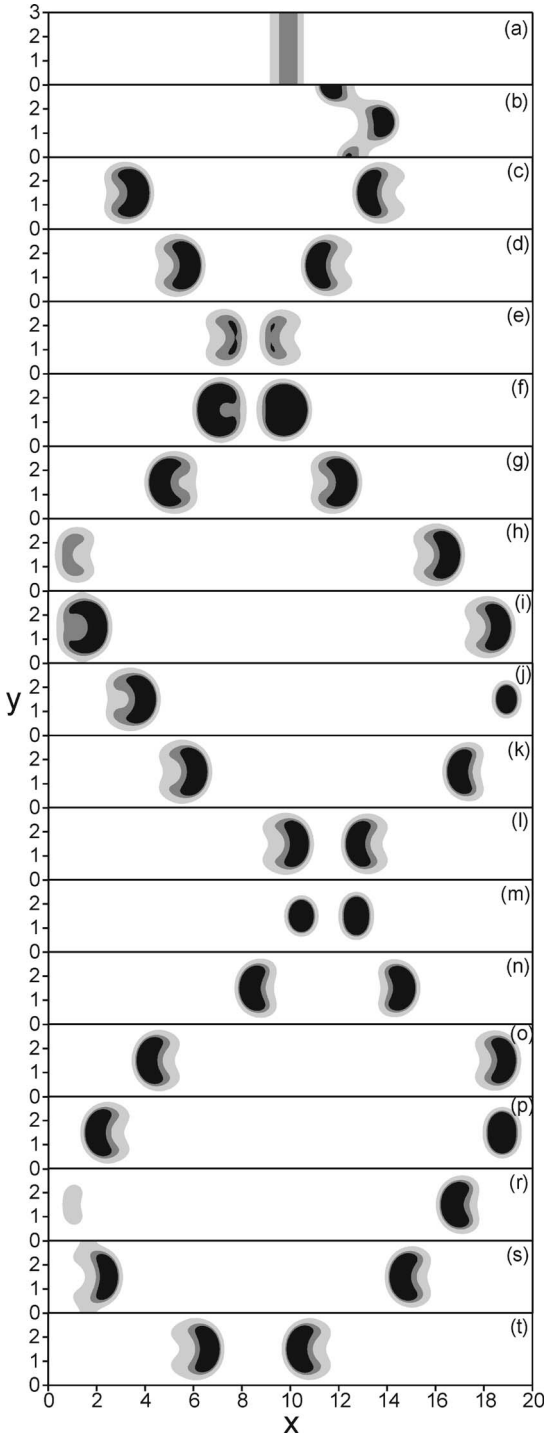

FIG. 4. Solutions to Eqs. (3), (4b), and (5) for $D=4.5$ and $\mathrm{pert}(x,y)=0.02$ at $(x,y)=(10.08,3)$ and 0 on all other points at $t=0$ (a), 0.336 (b), 1.664 (c), 1.672 (d), 1.68 (e), 1.688 (f), 1.696 (g), 1.712 (h), 1.72 (i), 1.728 (j), 1.736 (k), 1.752 (l), 1.76 (m), 1.768 (n), 1.784 (o), 1.792 (p), 1.8 (r), 1.808 (s), and 1.824 (t). The remaining parameter values are the same as in Fig. 2.

this paper, L'_{cr} is approximately equal to 6. For L greater than L'_{cr} the initially symmetric pattern becomes unstable, which results in the symmetry being lost. After complicated evolution the system approaches another attractor, which may be a curved impulse reflecting from the vertical walls or a homogenous stationary state. During this evolution many reflections, splitting, and decays of the impulses of excitation occur. For example at $L=6$ the system approaches a homogeneous stationary state. It is noteworthy that the homogeneous stationary state is approached if the initial conditions [Eq. (4a)] are asymmetrically perturbed at one point of the numerical grid. A curved traveling impulse reflecting from the vertical boundaries is obtained for $L=11$. It is noteworthy that propagation of the impulses along the diagonal also occurs in rectangular domains if the difference between L_x and L_y is small.

Figure 4(a) shows the evolution of the planar impulse perturbed at one point of the rectangular domain ($L_x > L_y$). For an appropriate initial perturbation $\mathrm{pert}(x,y)$ [Eq. (4b)], the impulse splits, creating two counterpropagating curved impulses [see Fig. 4(b)]. The subsequent evolution is very complex. The impulses split and collide with the boundaries or with one another. Consequently, they reflect from the boundaries and each other or occasionally disappear. After a long transient period between Figs. 4(b) and 4(c) (not shown here), a stable asymptotic pattern consisting of two curved traveling impulses is formed. The impulses periodically reflect from the boundaries and each other. One can see in Figs. 4(d) and 4(t) that the system is nearly in the same phase. The number of asymptotically reflecting impulses depends on the initial perturbation. Therefore, for a given system size different asymptotic patterns, which contain different numbers of reflecting impulses, may coexist.

IV. CONCLUSIONS

The problem of the existence of asymptotic stable periodic spatiotemporal 2D patterns in an activator-depleted reactant reaction-diffusion system is considered for the case when this system has a 1D asymptotically stable solution in the form of the traveling impulse periodically reflecting from impermeable boundaries. This problem is not trivial, because an asymptotically stable solution in an nD system may be unstable in $(n+1)D$ systems. The reflection of the traveling impulse in a 1D system does not automatically guarantee the reflection of the planar impulse in 2D systems. The planar traveling impulse shown in Fig. 4(a) is unstable although the 1D impulse shown in Fig. 2(a) is stable. Moreover, the almost circular traveling impulse generated in the square domain shown in Fig. 3(b) is unstable and transforms to a periodically changing impulse which is symmetric with respect to the diagonal. The breaking of the diagonal symmetry is the other problem that appears in evolutions of 2D patterns in square domains. Therefore, the reflection of the traveling impulse in 2D systems in our model is a much more complex phenomenon than the front wave reflecting with a single angle observed in the Belousov-Zhabotinsky reaction [11] and the specular reflection of the packet wave observed in the modified Brusselator model [9]. The shape of the traveling impulse after the reflection is different from that before the reflection.

Two types of stable periodic spatiotemporal patterns are presented. One of them has the form of a traveling impulse

propagating along the diagonal and reflecting from the boundaries of the square spatial domain (see Fig. 3). The other one consists of two curved traveling impulses reflecting from the boundaries and from each other in a rectangular spatial domain (see Fig. 4).

The model presented in this paper is the simplest one in which a traveling impulse and its reflection is possible. It describes the one-side-fed, unstirred reactor in which only one elementary reaction occurs in the gaseous phase with Lewis number less than 1. It is excitable and has only one stable homogeneous stationary state. The exponential dependence of the rate reaction constant on the temperature is necessary condition for the excitability of the system. The excitability has been observed in various two-variable RD systems like the FitzHugh-Nagumo [17,18], the Gray-Scott [19], and the Rovinsky-Zhabotinsky [20] models, the model based on the catalytic (enzymatic) reaction inhibited by the excess of the reactant and the product [21], and others. The results obtained for the model described in this paper may qualitatively correspond to results for other excitable activator-inhibitor RD systems in which the diffusion coefficient of the inhibitor is greater than the diffusion coefficient of the activator. The periodic spatiotemporal patterns described in the present paper increase the number of asymptotic patterns that have been observed in 2D RD systems. We hope that our paper will be helpful in observing them in experiments. Excitable systems such as chlorite-tetrathionate in polyacrylamide gel [22] and chlorite dioxide–iodine–malonic acid in polyvinyl alcohol [23] seem to satisfy the necessary conditions for the ratio of the activator and inhibitor diffusion coefficients.

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